ORGIA INSTITUTE OF TECHNOLOGY OFFICE OF CONTRACT ADMINISTRATION

PROJECT ADMINISTRATION DATA SHEET

[X] ORIGINAL [ ] REVISION NO. _____

ect No./(Center No.) E-20-G11 R6216-0A0 GTRC/~~GIT~~ DATE 1 / 26 / 87

ect Director: Dr. F. M. Saunders MISC School/~~XXX~~ Civil Engineering

nsor: American Water Works Association Research Foundation (AWWARF)

ement No.: Contract No. 213-86 ~~12/30/89~~ 4/30/88

ird Period: From 10/1/86 To ~~9/30/87~~ 12/30/89 (Performance) ~~9/30/87~~ 4/30/88 Reports

nsor Amount:

	New With This Change	Total to Date
Contract Value: $		$ 22,250
Funded: $		$ 22,250

Sharing No./(Center No.) _____ Cost Sharing: $ _____

: Critical Evaluation of Pilot and Full-Scale Coagulant Rocovery Systems

INISTRATIVE DATA OCA Contact E. Faith Gleason x. 4820

ponsor Technical Contact:

Nancy McTigue, Project Officer
AWWA Research Foundation
6666 West Quincey Avenue
Denver, CO 80235
(303)794-7711

2) Sponsor Issuing Office:

ıry Security Classification: _____ ONR Resident Rep. is ACO: _____ Yes X No

Company/Industrial Proprietary: N/A Defense Priority Rating: _____

RICTIONS

ttached _____ Supplemental Information Sheet for Additional Requirements.

l: Foreign travel must have prior approval — Contact OCA in each case. Domestic travel requires sponsor approval where total will exceed greater of $500 or 125% of approved proposal budget category.

ıment: Title vests with (none proposed)

MENTS:

ES TO: SPONSOR'S I.D. NO. 02,500,017,87,001

t Director	Procurement/GTRI Supply Services	GTRC
rch Administrative Network	Research Security Services	Library
rch Property Management	Contract Support Div.(OCA)(2)	Project File
	Research Communications	Other

GEORGIA INSTITUTE OF TECHNOLOGY
OFFICE OF CONTRACT ADMINISTRATION

NOTICE OF PROJECT CLOSEOUT

Closeout Notice Date 02/20/91

ect No. E-20-G11_____ Center No. R6216-0A0_____

ect Director SAUNDERS F M_____ School/Lab CIVIL ENGR____

sor AMER WATER WORKS ASSO RES FDN/DENVER, CO_____

ract/Grant No. 213-86_____ Contract Entity GTRC

e Contract No. _____

e CRITICAL EVALUATION OF PILOT AND FULL-SCALE COAGULANT RECOVERY SYSTEMS.__

ctive Completion Date 900101 (Performance) 900101 (Reports)

---

eout Actions Required:	Y/N	Date Submitted
inal Invoice or Copy of Final Invoice	Y	_____
inal Report of Inventions and/or Subcontracts	Y	_____
overnment Property Inventory & Related Certificate	N	_____
lassified Material Certificate	N	_____
elease and Assignment	N	_____
ther _____	N	_____

omments_____

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oject Under Main Project No. _____

nues Project No. _____

ibution Required:

roject Director	Y
dministrative Network Representative	Y
TRI Accounting/Grants and Contracts	Y
rocurement/Supply Services	Y
esearch Property Managment	Y
esearch Security Services	N
eports Coordinator (OCA)	Y
TRC	Y
roject File	Y
ther _____	N
_____	N

Final Patent Questionnaire sent to PDPI.

E-20-G-11

Georgia Institute of Technology

A UNIT OF THE UNIVERSITY SYSTEM OF GEORGIA

ATLANTA, GEORGIA 30332

SCHOOL OF
CIVIL ENGINEERING

TELEX: 542507 GTRC OCA ATL

TELEPHONE
(404) 894- 2265

26 June 1987

Ms. Nancy E. McTigue
AWWARF
6666 W. Quincy Avenue
Denver, CO 80235

Subject: Quarterly Report for AWWARF Project:
"Critical Evaluation of Pilot- and
Full-Scale Coagulant Recovery Systems"

Dear Ms. McTigue:

After numerous delays in obtaining final approval and in initiation of the project, progress is being made on the subject project. An Advisory Group has been established to assist in (i) identification of plants which have or are practicing or examining coagulant recovery; (ii) acquisition of unpublished data, operational records and engineering reports on coagulant recovery through consulting firms or water treatment plants; and (iii) critical review of published reports. The current members of this Advisory Group are listed in an appended table.

A computer-based literature search has been conducted to update our records of published research. The previous and newly acquired literature citations are being reexamined to obtain more detailed information and data about coagulant quality, reaction kinetics and system economics.

To assist in identification of plants conducting coagulant recovery studies, the AWWA has been requested to publish the enclosed notice in the Journal, Opflow and Mainstream. The notice was also used to request information on the quality of virgin coagulants, for use in comparison with that of recovered coagulants, and on the heavy-metal and organic composition of aluminum- and iron-sludges. At the recent AWWA conference, a brief poster presentation was made to inform the industry of the project and to request information, as seen in the enclosed illustrations.

Information regarding the level of inorganic and organic contamination (i.e., current SDWA contaminants) of conventional virgin coagulants has been requested from chemical suppliers. They, however, have been hesitant to supply these data. Mr. Rich Karlin of AWWA has been contacted to obtain information regarding the NSF/AWWARF/ASDWA/COSHEM Drinking Water Additives program and data available on coagulant quality.

One site visit has been made to a facility conducting plant-scale studies on coagulant recovery and visits to three additional locations are being scheduled. In depth analysis of the coagulant-recovery experiences at Jersey City are being pursued so as to establish definitive reasons for why this facility was not successful in implementation of coagulant recovery. Finally, contacts in England and Japan have been identified and are being contacted regarding coagulant recovery experiences in these countries.

With respect to progress during the coming quarter, a graduate research assistant has been hired to pursue remaining issues and to assist in development of a final report.

Sincerely,

F. Michael Saunders

FMS/hb
cc: Advisory Group

bcc: OCA
J. E. Fitzgerald

COAGULANT RECOVERY ADVISORY GROUP

AWWA Research Foundation Project:
"Critical Evaluation of Pilot- and Full-Scale
Coagulant Recovery Systems"

Project Director

F. Michael Saunders
Environmental Engineering
Georgia Institute of Technology
Atlanta, GA 30332-0512
(404) 894-2265

Project Officer

Nancy E. McTigue
AWWARF
6666 W. Quincy Avenue
Denver, CO 80235
(303) 794-7711

ADVISORY GROUP

Robert B. Rivers, Director
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236 Forsyth St., S.W.
Atlanta, GA 30303
(404) 658-6580

Carl Camp, Plant Superintendent*
Athens Water Plant
Athens Utility Board
Athens, TN 37303
(615) 745-6273

David Cornwell, President
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Jordan Jones and Goulding
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Atlanta, GA 30340
(404) 455-8555

Terry Rolan
City of Durham
Department of Water Resources
Durham, NC 27701

Billy G. Turner, General Manager/
Director
Spartanburg Water System
P. O. Box 251
Spartanburg, SC 29304
(803) 582-6375

*pending approval by City

-COAGULATION OF SURFACE WATERS-
ASSISTANCE OF WATER INDUSTRY NEEDED

The AWWARF is sponsoring a study to develop a critical assessment of coagulant recovery systems that have been or can be employed in the water treatment industry. The assistance of the industry is needed.

In this study, recovery of aluminum (Al) or iron (Fe) coagulants by direct extraction under acidic or alkaline conditions is being examined. Information and data are needed from individuals at water treatment plants, researchers, consultants and chemical manufacturers and suppliers in the following areas:

- Identification of plants which have conducted laboratory-, pilot- and full-scale studies of coagulant recovery, regardless of depth of studies;
- Case histories, results or raw data resulting from above studies;
- Data indicating concentrations of Ag, As, Ba, Cd, Cr, Hg, Se or any other inorganic or organic contaminants in aluminum or iron coagulants or coagulant aids used in water treatment plants;
- Data indicating concentrations of inorganic and organic contaminants in thickened sludge suspensions produced in coagulation.

Individuals having information in any of these areas are requested to contact the principal investigator, Dr. F. Michael Saunders at (404)894-2265 Environmental Engineering, Georgia Institute of Technology, Atlanta, GA 30332-0512).

CRITICAL EVALUATION OF PILOT- AND FULL-SCALE COAGULANT RECOVERY SYSTEMS

★★★★

F. Michael Saunders

Georgia Institute of Technology
Atlanta GA

1. OBJECTIVE

- **Develop a critical review of coagulant recovery processes for surface water plants with emphasis on:**

  *** recovered-coagulant quality and utility relative to virgin coagulants;**

  *** full-scale systems implemented and reasons for successes and failures;**

  *** impact of recovered coagulant on water quality;**

  *** utility of recovered coagulant for non-potable uses;**

  *** handling, treatment, and disposal of extracted sludges.**

2. APPROACH

- **Assemble and review available data and reports on lab-, pilot-, and full-scale coagulant-recovery studies**
- **Update coagulant-recovery literature**
- **Identify current and emerging standards on coagulant chemicals**
- **Identify contaminant chemicals in current coagulants and sludges**
- **Identify composition, quality, and utility of recovered coagulants**
- **Identify impact on sludge handling, treatment and disposal**
- **Assess costs of recovery systems**
- **Identify advantages and disadvantages of recovery systems**
- **Develop overall critical assessment of coagulant-recovery systems**

3. COAGULANT RECOVERY SYSTEMS

ALUMINUM and IRON

- **DIRECT RECOVERY**

  Acidic Extraction

  Alkaline Extraction

- **INDIRECT RECOVERY**

  Solvent Extraction

4. ALUMINUM AND IRON RECOVERY

Typical Extraction Process

- **Rapid mix of thickened sludge and acid**
- **Reaction period for extraction to proceed**
  (2 Al(OH)3 + 3 H2(SO4) --→
  Al2(SO4)3 + 6 H20)
- **Separation of suspended solids and coagulant solution by gravity settling or filtration**
- **Recycle of recovered coagulant**
- **Dewater and dispose of residual sludge**

5. PILOT- and FULL-SCALE STUDIES

DIRECT COAGULANT RECOVERY

- **ALUMINUM**

  Tampa FL - 1960
  Japan - 1965-1972
  Oakland CA - 1973
  Buffalo NY - 1974
  Jersey City NJ - 1974
  Atlanta GA - 1984-1987
  Durham NC - 1985-1987

- **IRON**

  Athens TN - 1984-1987

6. ADVANTAGES OF COAGULANT RECOVERY

- **Minimize quantities and costs of virgin coagulants**
- **Enhanced coagulation due to polymeric forms of metal ions and presence of other extracted coagulants**
- **Reduced sludge volume, mass and disposal costs**
- **Enhanced dewaterability of residual sludge by removal of gelatinous metal hydroxides**

7. DISADVANTAGES OF COAGULANT RECOVERY

- **Increased handling and storage of extractant chemicals and recovered coagulant**
- **Increased costs for coagulant aids (e.g.,lime, PAC, Cl2, KMnO4)**
- **Contamination of coagulant slurry with extracted inorganic and organic compounds (e.g., Mn, Fe, TOC, Color)**
- **Increased complexity of sludge handling, treatment and disposal systems**

8.PROJECT TIMETABLE

- **Project Initiated..... January 1987**

- **Draft Completion Report..... October 1987**

- **Final Report..... December 1987**

9. ASSISTANCE NEEDED from WATER TREATMENT INDUSTRY

- **Identification of plants which have conducted lab-, pilot-, or full-scale coagulant-recovery studies**

- **Case histories of coagulant-recovery studies and installations including cost data for extraction and sludge disposal systems**

- **Quantitative characterization of trace contaminants in :**
  virgin coagulants;
  recovered coagulants;
  coagulant-aids; and
  sludges.

- **Identification of local, state and federal regulations regarding use of recovered coagulants**

PLEASE LEAVE NAME AND ADDRESS IF YOU CAN PROVIDE ASSISTANCE WITH ABOVE

Environmental Engineering
School of Civil Engineering
Georgia Institute of Technology
Atlanta, Georgia 30332
(404) 894-2265

MEMORANDUM

Date: 14 DEC '87

To: INA LASHLEY

From: FM SAUNDERS

Subject: E20 G11

The attached letter from RJ Karlin is in response to a request of 1 October in a quarterly report. Please make this project extended accordingly.

cc. T. Fennell

Telex: 45-0895 AWWA DVR
Cable: WATERINFO

AWWA Research Foundation | 6666 West Quincy Avenue | Denver, CO 80235 | 303 794-7711

December 10, 1987

F. Michael Saunders, Ph.D., P.E.
Associate Professor
School of Civil Engineering
Georgia Institute of Technology
Atlanta, GA 30332

Dear Dr. Saunders:

This letter will serve as amendment to Contract 213-86 between the AWWA Research Foundation and Georgia Institute of Technology. The project period will now end on April 30, 1988 with the draft final report due sixty days prior to allow for PAC review and possible revision.

A quarterly report will be due on February 1, 1988. All other terms of the contract will still apply. Total funding from the AWWA Research Foundation remains at $22,250.00.

Sincerely yours,

Richard J. Karlin, P.E.
Director
Research Management Division

RJK:mas:213

Sorry for the delayed response — late

Georgia Institute of Technology
A UNIT OF THE UNIVERSITY SYSTEM OF GEORGIA
ATLANTA, GEORGIA 30332

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(404) 894-2265

1 October 1987

Ms. Nancy E. McTigue
AWWARF
6666 W. Quincy Avenue
Denver, CO 80235

SUBJECT: Quarterly Report for AWWARF Project -
"Critical Evaluation of Pilot- and Full-Scale
Coagulant Recovery Systems"

Dear Nancy:

Considerable progress has been made during the past quarter. Information has been obtained from the chief design engineer regarding the implementation of the coagulant recovery system at Jersey City and the reasons for why the much-heralded coagulant-recovery facility was never actually placed in operation. This new information will place this system, frequently referenced as a facility at which coagulant recovery was a failure, in a new perspective.

A cooperative relationship has been established with the Athens Utility Board of Athens, Tennessee as of 4 August 1987. The Board operates a municipal surface-water treatment plant which uses iron salts as a primary coagulant. A system for acidic extraction of its coagulant sludge was implemented in 1985 and has been used intermittently for the past two years. Operational data for the batch extractions of the iron sludges and for the total plant for periods during which the recovered coagulant were being used have been provided. These data have been evaluated with respect to recovered-coagulant properties and quality; impact on the performance of filtration systems and on finished-water quality; and impact on operational costs with and without coagulant recovery. The assessment to date has indicated that intermittent use of recovered coagulant has had no negative impact on effluent quality (including data for system THMFP and MCL parameters); slightly decreases the length of filter runs; dramatically improves sludge treatment properties and minimizes sludge disposal costs; and results in monetary savings in the purchase of coagulant and coagulant aids. These data have been summarized and are near completion. Prior to distribution they must be approved by the Athens Utility Board.

As a result of the publication by AWWA of requests for information on coagulant recovery systems, numerous responses have been received. The majority of these responses have focused on the quality of virgin coagulants and chemical composition of sludge solids.

After considerable correspondence with Japanese researchers, contact has been made with Dr. Yasumota Magara, Director of the Department of Sanitary Engineering of the Institute of Public Health in Tokyo. He has just recently (21 September 1987) provided me with considerable information regarding coagulant recovery in Japan, including an extensive (293 pp) report on optimization of water treatment and sludge disposal, including in-depth coverage of coagulant recovery. This information will serve to document the long-term Japanese experience with full-scale, coagulant-recovery systems. Dr. Magara, in addition, has identified six (6) plants which are currently practicing coagulant recovery using acidic extraction processes. Further efforts are being made to contact individuals at these plants to collect operational data for these systems. Placed in the context of only one, full-scale plant (i.e., Athens, TN) currently practicing coagulant recovery in the USA, acquisition of further documentation from Japan is critical so as to provide a comprehensive critical assessment of full-scale coagulant recovery systems. Although the actual period of operation is unknown at this point, these six (6) plants, as well as others previously, have practiced coagulant recovery for periods in excess of five years, making acquisition of this information even more critical.

The current schedule for completion of the project requires that a draft final report be submitted on 30 October 1987 and that the final report be completed as of 30 December 1987. The administrative delays associated with acquisition and review of data from full-scale plants in the USA and the recent acquisition of extensive data from Japan require that a request be made to extend the final submission date for the project report. In addition, the final report is to be coordinated with the AWWARF sludge report, which is in the publication process, and is not currently available. Therefore, it is requested that a 4-month, no-cost extension be granted to allow for acquisition of the critical Japanese data and finalize the project report. I am confident that with the data currently available and the recent establishment of a Japanese technical contact that the requested extension is absolutely necessary to assure that a full assessment is made of full-scale coagulant recovery systems. Review of the previously unavailable Japanese data should provide the water supply industry in the USA with a revealing and valuable documentation of the potential for implementation of coagulant recovery. Your approval of this request and the support of Kim Fox and Charlie O'Melia will be appreciated.

Sincerely,

F. Michael Saunders
Project Director

MS:es

c: K. Fox
C. O'Melia
P. Heitmueller
T. Fennell

Georgia Institute of Technology

A UNIT OF THE UNIVERSITY SYSTEM OF GEORGIA

ATLANTA, GEORGIA 30332

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2 March 1988

Mr. Richard J. Karlin
AWWA Research Foundation
6666 W. Quincy Avenue
Denver, CO 80235

Subject: Critical Evaluation of Pilot- and
Full-Scale Coagulant Recovery Systems

Dear Rick:

As we discussed yesterday, the draft of the final project report is being delayed in anticipation of receiving informaton from eight full-scale plants in Japan using coagulant recovery systems. After some initial confusion, preliminary information was provided by Dr. Yasumoto Magara of the Japanese Institute of Public Health. As indicated in an attached letter, additional information was requested in November 1987 and again in February. A recent telex message from an assistant to Dr. Magara, indicates information on six plants has been collected and information on the two remaining plants will be available soon.

Inclusion of long-term operating data for full-scale Japanese plants in the report is highly desirable and is of great interest to professionals in this country. It is requested that the project be extended to allow for evaluation of the forthcoming data and its inclusion in the report. As we agreed yesterday, I will alert you to my receipt of the Japanese data and will provide you with an assessment of the time required to review and evaluate the data for inclusion in the final project report. At that time a new date for submission of the final report can be established.

Your assistance with this matter is greatly appreciated.

Sincerely,

F. Michael Saunders

FMS/hb
Enclosure
cc: K. Fox
C. O'Melia
R. Rivers

Georgia Institute of Technology
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23 February 1988

Dr. Yasumoto Magara, Director
Department of Sanitary Engineering
The Institute of Public Health
6-1, Shirokanedai 4 chrome
Minato-ku
TOKYO 108 JAPAN

SUBJECT: Coagulant Recovery in Japanese Water Treatment Plants

Dear Dr. Magara:

I have learned through several AWWA and EPA publications that you were in the USA several months ago and hope your trip was a successful one. My only regret was that I was unable to meet with you to discuss water treatment practices in Japan.

On 30 November 1987, I forwarded a lengthy letter to you regarding coagulant recovery practices in Japan. I am confident that your schedule was full, and even overflowing, with tasks to be addressed by you and the Department and I apologize for adding to this with my letter. However, my current project with the AWWARF would be incomplete without inclusion of the full-scale experiences with coagulant recovery in Japan. Due to problems with international mail, you may not have received the letter of 30 November so I have enclosed a copy for your reference.

I am eager to learn if you will be able to assist me in acquisition of the data and information requested, as are the members of my Advisory Committee (Mr. Kim Fox of EPA, Mr. Rick Carlin of AWWARF and Dr. Charles O'Melia of The Johns Hopkins University). As indicated previously, collaboration with you in the development of a publication or report on this topic is a definite possibility. If possible, please indicate by return mail if we will be able to collaborate on this matter. If you have been able to identify a few initial sources of information, receipt of these will be appreciated. I recognize this is a major imposition but also am keenly aware of the major professional contribution which can be made nationally and internationally with your assistance in this area.

Your kind and continuing assistance will be greatly appreciated.

Sincerely,

F. Michael Saunders, Ph.D., P.E.

Correspondence Address:
Environmental Engineering
Georgia Institute of Technology
Atlanta, GA 30332-0512 USA

Georgia Institute of Technology
A UNIT OF THE UNIVERSITY SYSTEM OF GEORGIA
ATLANTA, GEORGIA 30332

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30 November 1987

Dr. Yasumoto Magara, Director
Department of Sanitary Engineering
The Institute of Public Health
6-1, Shirokanedai 4 chrome
Minato-ku
TOKYO 108 JAPAN

Dear Dr. Magara:

Thank you very much for your letter of 21 September 1987 and the reports and publications which were included. With the assistance of a student who can read Japanese, I have been able to make use of the data provided. I, however, would like to obtain more operational and performance information on systems using coagulant recovery.

In your letter you indicated that no acid-leaching processes, or coagulant recovery systems, have been included in water treatment plants which have been built since about 1977. I understand that the reasons you indicated for discontinuation of coagulant recovery systems included: (i) accumulation of heavy metals and THM precursors in recovered coagulants; (ii) management and storage of large quantities of dilute recovered coagulant; and (iii) increased safety requirements associated with handling sulfuric acid. These results are not unexpected in view of my studies of coagulant recovery systems. However, we in the United States have very limited full-scale experience with coagulant recovery systems, making it difficult to document and quantify these problems. This then leads to the reason for a request for your assistance and potential collaboration.

The sponsored study I am conducting for the American Water Works Association Research Foundation (AWWARF), and the U.S. Environmental Protection Agency as a cosponsor, is to make a critical assessment of full-scale and pilot-scale coagulant recovery systems and to definitively identify the advantages and disadvantages of the process when applied to full-scale plants. Because Japanese water treatment works have such extensive experience in coagulant recovery, it is extremely necessary that Japanese experiences be included in our study, if the final report is to be complete and comprehensive.

In the information previously forwarded, you indicated on a summary table that eight water treatment plants located in Saitama-ken (Okubo and Mouwa), Tokyo, Yokohama-City, Itan-City, Seto-Chou, Hiroshima-City and Fukui-City (see enclosed list for plant names in Japanese and English) currently use acid-leaching processes to recover coagulants. It would be most helpful if you could assist me in obtaining plant operational and performance data from all of these plants.

The detailed information needed from all, if possible, of these plants includes:

(i) sources and biological, chemical and physical characteristics of raw water supplies being treated by each plant;

(ii) a description of the unit operations and processes, including typical hydraulic loadings and chemical application rates, for the water-treatment system and the sludge-treatment system at each plant;

(iii) operational procedures and data for the acid leaching (i.e., coagulant recovery) of sludges including: typical sludge suspended solids concentrations; type and quantity of acid added; pH of acidified sludge and time of reaction; means of recovery of soluble coagulant and separation of residual suspended solids; typical chemical characteristics of recovered coagulant with emphasis on pH, Al^{+3} concentration, Fe and Mn concentrations, trace heavy metals and other metals, trace organics, color and THMFP; and volume of coagulant storage facilities and frequency with which recovered coagulant is produced;

(iv) information on the accumulation of contaminants in recovered coagulants and how this is controlled;

(v) chemical additives and the quantities required by the use of recovered coagulants (e.g., lime or NaOH for pH adjustment and oxidants for manganese oxidation) and a comparison of quantities of these chemicals used in conjunction with the use of virgin coagulants (i.e., during periods when recovered coagulant is not available), as well as an estimate of cost savings associated with coagulant recovery (i.e., costs of acid and other chemical additives compared with costs for a virgin alum system);

(vi) level of contamination of treated water attributed to the use of a recovered coagulant; and

(vii) the chemical characteristics of virgin alum or other coagulant used in water treatment, as well as the source of alum (e.g., is bauxite the source of alum?) and the costs and availability of alum.

ata similar to these have been obtained for one USA facility [Athens, TN hich uses iron (Fe^{+2}) as a coagulant] from plant operational data provided y plant personnel. From my experience, monthly plant operational records, n conjunction with a firm understanding of the treatment plant, can provide n excellent basis for initiation of such an investigation. These data, and ıose from annual summaries and water-quality reports, may provide us with ıe necessary information to initiate our studies of Japanese coagulant

recovery systems. Your assistance in obtaining this information from individual plants will be appreciated. If appropriate, please provide me with the name of a key individual at each plant whom I might contact to obtain this information. I am, however, concerned that I may not be able to contact someone at each plant who has the same excellent command of English as you do. Therefore, please suggest how I, with only a Japanese translator, can best obtain these data as rapidly as possible.

The success of this project focused on coagulant recovery will be assured if Japanese data can be used to document the advantages and disadvantages of the process. For example, from studies I have conducted in Atlanta, Georgia along the Chattahoochee River, laboratory data indicate that trace metals and trace organics will not present a problem for a coagulant recovery system. However, without full-scale experience it is impossible to firmly establish this point. Therefore, your collaboration and assistance in identifying informational sources which can clearly document the impact of coagulant recovery on full-scale water works will be greatly appreciated.

I will eagerly await your response on the proposed direction to be taken and any assistance you can provide.

Sincerely,

F. Michael Saunders, Ph.D., P.E.

Address Correspondence to:

Environmental Engineering
Georgia Institute of Technology
Atlanta, GA 30332-0512 USA

'MS/hb

TRANSLATION OF NAMES OF SPONSORS AND WATERWORKS PLANT
WHICH USE ACID-LEACHING OF COAGULATION SLUDGE SOLIDS
TO RECOVER OR RELCAIM COAGULANT CHEMICALS OR REDUCE
SLUDGE QUANTITIES

ENGLISH		JAPANESE	
Sponsor	Plant Name		
Saitama-ken	Okubo	埼玉県	大久保
Saitama-ken	Shouwa	埼玉県	庄和
Tokyo	Misono	東京都	三園
Yokohama-City	Kosuzume	横浜市	小雀
Itan-City	Senzou	伊丹市	千僧
Seto-Chou	Ouchi	瀬戸市	大内
Hiroshima-City	Ryokui	広島市	緑井
Fukui-City	Nakazuhara	福山市	中津原

mail

GE # 11
J 2

)CA ATL

)7INSTPH J
MICHAEL SAUNDERS
)UR REQUEST ON COAGULANT RECOVERY IN JAPAN THE RELEVANT
IATION FOR EIGHT PLANTS PRACTICING THAT TREATMENT ARE BEING
:TED. NOW ANSWERS FROM SIX PLANTS HAVE BEEN RECEIVED AND ALL
ILL BE AVAILABLE IN SOME WEEKS.
IGH DR MAGARA IS ON DUTY TRIP. IT MAY BE POSSIBLE TO SEND
ITA OF SIX PLANTS ON REQUEST.

NE FOR MAGARA
UBLIC HEALTH TOKYO

CA ATL

LY FROM TELEX I OR II (TWX) DIAL 100 FROM EASYLINK USE /WUW.
27 FEB/26/1988

ON 120 SECS LISTED 04:35/02-206-01-00

OSA COMMUNICATIONS DESK

Date 2/26-88 Time 9:00 Operator S. Mill

TRANSMITTED: ____
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TWX
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Night Letter ____
Mailgram ____
Telegram ____
FAX ____

RECEIVED: ____
Person notified: ____
_____
Time: _____ Date: ____
NOTE:

Original to: Dr. Saunders
Copies to. _____
_____
_____

Georgia Institute of Technology
A UNIT OF THE UNIVERSITY SYSTEM OF GEORGIA
ATLANTA, GEORGIA 30332

SCHOOL OF
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1 October 1987

Ms. Nancy E. McTigue
AWWARF
6666 W. Quincy Avenue
Denver, CO 80235

SUBJECT: Quarterly Report for AWWARF Project -
"Critical Evaluation of Pilot- and Full-Scale
Coagulant Recovery Systems"

Dear Nancy:

Considerable progress has been made during the past quarter. Information has been obtained from the chief design engineer regarding the implementation of the coagulant recovery system at Jersey City and the reasons for why the much-heralded coagulant-recovery facility was never actually placed in operation. This new information will place this system, frequently referenced as a facility at which coagulant recovery was a failure, in a new perspective.

A cooperative relationship has been established with the Athens Utility Board of Athens, Tennessee as of 4 August 1987. The Board operates a municipal surface-water treatment plant which uses iron salts as a primary coagulant. A system for acidic extraction of its coagulant sludge was implemented in 1985 and has been used intermittently for the past two years. Operational data for the batch extractions of the iron sludges and for the total plant for periods during which the recovered coagulant were being used have been provided. These data have been evaluated with respect to recovered-coagulant properties and quality; impact on the performance of filtration systems and on finished-water quality; and impact on operational costs with and without coagulant recovery. The assessment to date has indicated that intermittent use of recovered coagulant has had no negative impact on effluent quality (including data for system THMFP and MCL parameters); slightly decreases the length of filter runs; dramatically improves sludge treatment properties and minimizes sludge disposal costs; and results in monetary savings in the purchase of coagulant and coagulant aids. These data have been summarized and are near completion. Prior to distribution they must be approved by the Athens Utility Board.

As a result of the publication by AWWA of requests for information on coagulant recovery systems, numerous responses have been received. The majority of these responses have focused on the quality of virgin coagulants and chemical composition of sludge solids.

After considerable correspondence with Japanese researchers, contact has been made with Dr. Yasumota Magara, Director of the Department of Sanitary Engineering of the Institute of Public Health in Tokyo. He has just recently (21 September 1987) provided me with considerable information regarding coagulant recovery in Japan, including an extensive (293 pp) report on optimization of water treatment and sludge disposal, including in-depth coverage of coagulant recovery. This information will serve to document the long-term Japanese experience with full-scale, coagulant-recovery systems. Dr. Magara, in addition, has identified six (6) plants which are currently practicing coagulant recovery using acidic extraction processes. Further efforts are being made to contact individuals at these plants to collect operational data for these systems. Placed in the context of only one, full-scale plant (i.e., Athens, TN) currently practicing coagulant recovery in the USA, acquisition of further documentation from Japan is critical so as to provide a comprehensive critical assessment of full-scale coagulant recovery systems. Although the actual period of operation is unknown at this point, these six (6) plants, as well as others previously, have practiced coagulant recovery for periods in excess of five years, making acquisition of this information even more critical.

The current schedule for completion of the project requires that a draft final report be submitted on 30 October 1987 and that the final report be completed as of 30 December 1987. The administrative delays associated with acquisition and review of data from full-scale plants in the USA and the recent acquisition of extensive data from Japan require that a request be made to extend the final submission date for the project report. In addition, the final report is to be coordinated with the AWWARF sludge report, which is in the publication process, and is not currently available. Therefore, it is requested that a 4-month, no-cost extension be granted to allow for acquisition of the critical Japanese data and finalize the project report. I am confident that with the data currently available and the recent establishment of a Japanese technical contact that the requested extension is absolutely necessary to assure that a full assessment is made of full-scale coagulant recovery systems. Review of the previously unavailable Japanese data should provide the water supply industry in the USA with a revealing and valuable documentation of the potential for implementation of coagulant recovery. Your approval of this request and the support of Kim Fox and Charlie O'Melia will be appreciated.

Sincerely,

F. Michael Saunders
Project Director

FMS:es

cc: K. Fox
C. O'Melia
P. Heitmueller
T. Fennell

COAGULANT RECOVERY: A CRITICAL ASSESSMENT

by

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April 1989

DISCLAIMER

This study was funded by the American Water Works Association Research Foundation (AWWARF). AWWARF assumes no responsibility for the content of the research study reported in this publication, or for the opinions or statements of fact expressed in the report. The mention of tradenames for commercial products does not represent or imply the approval or endorsement of AWWARF. This report is presented solely for informational purposes.

Although the research described in this document has been funded in part by the United States Environmental Protection Agency through a Cooperative Agreement, CR-811335-01, to AWWARF, it has not been subjected to Agency review and therefore does not necessarily reflect the views of the Agency and no official endorsement should be inferred.

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FOREWORD

This report is part of the on-going research program of the AWWA Research Foundation. The research described in the following pages was funded by the Foundation in behalf of its members and subscribers in particular and the water supply industry in general. Selected for funding by AWWARF's Board of Trustees, the project was identified as a practical, priority need of the industry. It is hoped that this publication will receive wide and serious attention and that its findings, conclusions, and recommendations will be applied in communities throughout the United States and Canada.

The Research Foundation was created by the water supply industry as its center for cooperative research and development. The Foundation itself does not conduct research; it functions as a planning and management agency, awarding contracts to other institutions, such as water utilities, universities, engineering firms, and other organizations. The scientific and technical expertise of the staff is further enhanced by industry volunteers who serve on Project Advisory Committees and on other standing committees and councils. An extensive planning process involves many hundreds of water professionals in the important task of keeping the Foundation's program responsive to the practical, operational needs of local utilities and to the general research and development needs of a progressive industry.

All aspects of water supply are served by AWWARF's research agenda: resources, treatment and operations, distribution and storage, water quality and analysis, economics and management. The ultimate purpose of this effort is to assist local water suppliers to provide the highest possible quality of water, economically and reliably. The Foundation's Trustees are pleased to offer this publication as contribution toward that end.

The disposal of water plant wastes continues to be a major problem for utilities of all sizes and reclamation of coagulant value can aid in reducing sludge quantities for disposal. This manual will be an invaluable resource for utility managers and design engineers by providing state-of-the-art information on all aspects of reclamation of aluminum and iron coagulants.

Jerome B. Gilbert
Chairman, Board of Trustees
AWWA Research Foundation

James F. Manwaring, P.E.
Executive Director
AWWA Research Foundation

EXECUTIVE SUMMARY

Coagulant Recovery: A Critical Assessment

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April 1989

AWWARF Order Number:

This document summarizes the current status of coagulant recovery technology, with a detailed critical discussion of the literature and evaluation and presentation of previously unpublished data on full-scale coagulant recovery operations in the United States and Japan. Operational and economical factors which impact implementation of coagulant recovery processes have been identified for water treatment sludges produced using aluminum and iron coagulants. The objective of this study was to provide the water utility industry with a comprehensive report on the water quality impacts and economic feasibility of coagulant recovery systems, as compared to conventional systems using commercial coagulants. The following is a summary of the topics described in this report.

CONCLUSIONS AND RECOMMENDATIONS

An overall summary of the conclusions and recommendations of the report are presented. Operational parameters impacting coagulant recovery, recovered-coagulant quality and coagulant utility are discussed. The treatment of residual sludge solids and overall economic analysis of coagulant recovery systems are also examined.

INTRODUCTION

Coagulation practices in potable water treatment are presented, including a general discussion of coagulation chemistry and typical coagulant application practices in water treatment. Commercial coagulant quality is discussed in the context of the Water Chemical Codex (National Research Council 1982) which has established guidelines for the quality of chemicals used in water treatment. The production of water treatment sludges from the use of commercial aluminum and iron coagulants is examined and the characteristics of typical water treatment sludges are discussed. The increasingly stringent regulations governing the disposal of water treatment plant sludges are discussed as they relate to the effectiveness of the coagulant recovery process in the conditioning of these sludges for final disposal.

ALUMINUM RECOVERY FROM WATER TREATMENT PLANT SLUDGES

Acidic and alkaline recovery of aluminum from water treatment sludges, resulting from the coagulation of raw waters with aluminum coagulants, is examined. Coagulant recovery process variables were defined and compared to theoretical predictions where possible. The quality of the recovered coagulants was examined with respect to recovered aluminum concentrations, as well as organic and inorganic trace contaminant concentrations. The utility of recovered coagulants is discussed, along with recommendations for areas requiring further research. Finally, the treatment of residual sludge solids following aluminum recovery and the volumetric and mass reductions in solids due to use of coagulant recovery processes are examined.

IRON RECOVERY FROM WATER TREATMENT PLANT SLUDGES

Recovery of iron from water treatment plant sludges resulting from the treatment of raw waters using iron coagulants was examined in the same manner as recovery of aluminum coagulants. Operation variables, recovered coagulant quality and utility and residual sludge solids treatment are discussed.

FULL-SCALE COAGULANT RECOVERY OPERATIONS

Full-scale coagulant recovery operations and testing in the United States and Japan are discussed in detail. Full-time operation at Athens, TN and in Japan are utilized to further investigate operational and economic considerations of coagulant recovery systems. Other investigators into full-scale implementation of coagulant recovery are discussed and three independent economic evaluations of capital, operating and maintenance costs are presented. The major variables impacting the operational and economical efficiency of coagulant recovery systems were investigated using the detailed data base obtained from full-scale investigations.

CONCLUSIONS AND RECOMMENDATIONS

Coagulant recovery processes for reclamation of aluminum and iron coagulants from water treatment plant sludges have been investigated in the United States since about 1960, resulting in full-scale design and implementation at a few sites. The water treatment plant at Athens, TN has utilized full-scale coagulant recovery since 1985 and is the only active full-scale facility in the United States. Virtually all wasted sludge solids in 1988 were conditioned with sulfuric acid for recovery of the primary ferric coagulant. Recovered coagulant was used to treat 35 percent of the water produced in 1988, providing an excellent data base for the investigation of coagulant recovery at a full-scale plant in the United States. In Japan, coagulant recovery has been used for nearly two decades to minimize sludge volume and mass for disposal and for the recovery of an effective coagulant for use in water treatment.

Based on the results of laboratory- and pilot-scale testing reported in the literature and information obtained in conjunction with detailed analysis of full-scale operations at the Athens, TN facility and nine full-scale installations in Japan, operational and economical implications resulting from the use of coagulant recovery systems have been identified. Coagulant recovery operational parameters, recovered coagulant quality and utility, residual sludge solids handling and economic considerations associated with construction and operation of coagulant recovery systems have been examined.

OPERATIONAL PARAMETERS IMPACTING COAGULANT RECOVERY

Acidic coagulant recovery has been investigated at the laboratory-, pilot- and full-scale levels, while alkaline recovery has received limited attention due primarily to inefficient extraction of primary coagulants. Sulfuric acid and hydrochloric acids have been investigated for use in acid recovery processes, with sulfuric acid being preferred due to its lower costs and equivalent, if not more efficient, extraction of the primary coagulant. The majority of this report is focused on sulfuric acid recovery of aluminum and iron coagulants produced in conjunction with coagulation of raw waters for the production of potable water.

Extraction of aluminum and iron coagulants from water treatment plant sludges with sulfuric acid can be controlled through use of stoichiometric acid additions based on dissolution of aluminum and iron. In most instances, acid doses were normalized to total coagulant-metal or recovered coagulant-metal concentrations and compared to stoichiometric predictions which were based on the coagulant-metal being present as $Al(OH)_3 \cdot 3H_2O$ or $Fe(OH)_3 \cdot 3H_2O$ in the sludges. The stoichiometric sulfuric acid demand based on dissolution of $Al(OH)_3 \cdot 3H_2O$ to Al^{3+} is 5.45 kg H_2SO_4/kg of Al, while the acid demand for dissolution of $Fe(OH)_3 \cdot 3H_2O$ to Fe^{3+} is 1.96 kg H_2SO_4/kg of Fe. However, some of the sludge metals present are in the form of oxides (e.g., $Al_2O_3 \cdot nH_2O$ and $Fe_2O_3 \cdot 3H_2O$) and have a different stoichiometric acid demand than the hydroxide species. Furthermore, these oxides require much more rigorous extraction conditions than the hydroxide species and may not be extractable under the conditions employed in coagulant recovery processes. Therefore, the use of stoichiometry as a relative indicator of acid addition can be misleading.

Other acid-demanding constituents in sludges also impact normalized acid doses. Due to dissolution of sludge heavy metals and other acid demanding constituents, sulfuric acid doses required for adequate extraction of aluminum and iron would be expected to be greater than that predicted stoichiometrically. Consequently, reported normalized acid doses have ranged from 1.0 to 2.0 times those predicted on a stoichiometric basis. Although the normalized acid requirement can be considered to be fairly constant for a given sludge, coagulant-metal concentrations, sludge solids content and other characteristics of raw sludges can change considerably with plant operations and raw water quality. Based on the above, use of acid addition as a process control parameter is difficult, as changes in sludge characteristics may have a dramatic impact.

Many investigators have evaluated aluminum and iron extraction from sludges based on suspension pH following acid addition. Although optimum pH values were found to be site specific, typical pH values ranged from approximately 2.0 to 3.0 and were found to remain fairly constant at a given site (Magara 1988a,b,e; Camp 1989). Based on the above considerations of stoichiometric acid additions and suspension pH values for process control parameters, suspension pH value has proven to be the most realistic parameter for full-

scale operation. The pH value is easily measured and has been utilized efficiently as a process control parameter in full-scale operations in the United States and Japan. However, acid addition is dependent on the target pH value which should be determined prior to full-scale implementation. It appears reasonable to assume that required acid additions will fall within the range of 1.0 to 1.5 times stoichiometric requirements and will typically result in suspension pH values in the range of 2.0 to 3.0.

The detention time for acid contact has also been identified as an important parameter in the acidic extraction of coagulants. It has been demonstrated that the extraction of coagulant metal in the coagulant recovery process reaches equilibrium in a period of less than 30 min. That is, the amount of extractable coagulant metal reached a maximum within 30 min of acidification, with no further extraction for time periods up to 60 min. Detention times employed for acid contact ranged from 10 min in full-scale operation in Japan (Westerhoff and Daly 1974; Goldman and Watson 1975) to 30-45 min during laboratory-scale testing in Atlanta (Saunders 1989). The time for acid contact can have a dramatic impact on the percent of total aluminum or iron extracted. Although experimental investigations should be conducted to determine an optimum contact time for a particular sludge, a period of 10 to 20 min appears to be adequate for maximum aluminum and iron dissolution.

QUALITY OF RECOVERED COAGULANTS

The quality of recovered coagulants has been evaluated primarily on the basis of the concentration of the primary coagulant metal (i.e., aluminum or iron) and the concentrations of impurities such as organic matter, manganese, trace metals and organic priority pollutants. The coagulant metal concentration will impact the effectiveness of recovered coagulants used in the production of potable waters, while contaminants are of concern since they may be recycled to the point of coagulant addition and impact required chemical additions and the resulting finished water quality.

Coagulant-Metal Concentrations in Recovered Coagulants

Aluminum concentrations in recovered coagulants ranged from 58 mg/L as Al

(Magara 1988a) to 3700 mg/L as Al (Gruninger 1975) in laboratory-, pilot- and full-scale testing of aluminum extraction. The percentage of total sludge aluminum extracted ranged from 60 percent (Isaac and Vahidi 1961) to 99 percent (Cornwell and Susan 1979) in laboratory- and pilot-scale testing, while Bishop et al. (1987) reported 79 percent and 74 percent recovery in two full-scale tests of coagulant recovery at Durham, NC. Recovered iron coagulant concentrations were similar to those in recovered aluminum coagulants. For example, recovered iron coagulant concentrations during full-scale operation at Athens, TN ranged from 2200 to 6400 mg/L as Fe (Camp 1989).

Aluminum and iron concentrations in recovered coagulants were found to be a function of raw sludge characteristics, quantity of acid addition, acidified suspension pH, and acid-contact detention time. However, another factor, the source of aluminum and iron contained in water treatment sludges (i.e., from the addition of coagulants or attributable to raw water solids) may control the maximum amount of coagulant metal extractable with stoichiometric sulfuric acid additions. For example, addition of an aluminum coagulant to a water results primarily in formation of aluminum hydroxides, which are quickly solubilized upon addition of acid to the sludge suspension. Aluminum associated with the raw water solids is typically in the form of aluminum oxides which are much more resistant to dissolution, requiring increased detention times and elevated temperatures for complete dissolution, similar to the conditions for extraction of aluminum from bauxite for production of commercial alum. When raw water solids loading is high compared to the solids produced from coagulant addition, aluminum oxides may represent a large portion of the total aluminum present in the sludge. These aluminum oxides may not be dissolved at typical pH values, temperatures and detention times employed in coagulant recovery processes. The same argument holds for the dissolution of iron oxides present in the raw water solids and controls the maximum amount of iron extractable from water treatment sludges in the coagulant recovery process. Therefore, 100 percent dissolution of coagulant metal in a water treatment sludge may not be attainable using sulfuric acid coagulant recovery. Furthermore, complete dissolution of total sludge metal may not be required to recover the mass of metal added as a coagulant in water treatment. Therefore, less than 100 percent coagulant metal dissolution can be expected for most water treatment sludges, but does not necessarily

indicate poor performance of the coagulant recovery process.

Solids separation is also a limiting factor in the maximum amount of coagulant that can be recovered. The portion of total coagulant metal solubilized by acid addition and the portion of liquid recoverable through solids separation in combination control the amount of coagulant recovered. For example, if 90 percent metal dissolution is obtained and 70 percent of the liquid volume is recovered, the coagulant-metal recovery would be 63 percent (i.e., 0.90 x 0.70 x 100) of the total coagulant metal in the sludge. Therefore, the percent of coagulant-metal dissolution and the degree of solids separation attainable will control the maximum amount of the primary coagulant which can be recovered.

Recovered Coagulant Contaminants

The Water Chemicals Codex (National Research Council 1982) has established Recommended Maximum Impurity Content (RMIC) values for water treatment chemicals. Specific analytical techniques are described for determination of these contaminant concentrations. However, these techniques have not been employed for determination of recovered coagulant contaminants. Consequently, comparison of contaminant concentrations reported in recovered coagulants to RMIC values established by the Water Chemicals Codex (National Research Council 1982) is a conservative one. That is, RMIC values are based on contaminant concentrations in solution following pH adjustment to a value of 6.0, whereas recovered coagulant contaminant concentrations have been determined based on total contaminant concentrations in the acidic solutions recovered. Therefore, recovered coagulant contaminants that are insoluble at a pH value of 6.0 but have not been precipitated and removed from solution by filtration, result in an artificially inflated contaminant concentration as compared to RMIC values. However, with this bias in mind, recovered coagulant contaminant concentrations were compared to RMIC values for a relative indication of recovered product quality.

Iron has been identified as a major contaminant in recovered aluminum coagulants, but an RMIC value has not been established for it. This metal, however, contributes to the coagulant value of recovered aluminum coagulants

since it will precipitate during coagulation and is of minor concern with respect to finished water quality.

Manganese has been identified as a troublesome contaminant in many recovered coagulants. Full-scale operational data obtained at Athens, TN indicates that manganese levels are in the range of 220 to 500 mg/L in recovered iron coagulants. Full-scale operational data indicate that oxidant demand may increase due to oxidation and precipitation of manganese. However, manganese concentrations in the finished water were not impacted by the use of recovered coagulants. Examination of full-scale operation in Japan indicated the possibility for build-up of manganese in recovered coagulants, but limited information was available on the impact on chemical application rates or finished water quality.

Extensive information on trace metals in recovered coagulants has been presented by Saunders (1989), Magara (1988b,g) and Bishop et al. (1987). Comparison of recovered coagulant trace metal contaminants to RMIC values indicated that chromium and lead concentrations in recovered coagulants are high and may exceed RMIC values (Saunders 1988; Bishop et al. 1987). Magara (1988b) reported high arsenic concentrations in recovered coagulants in Japan, but finished water arsenic concentrations were well below the Maximum Contaminant Levels. Other trace metals were present at concentrations substantially below specified RMIC values. Based on the conservative nature of these comparisons of total contaminant concentrations to RMIC values, however, it appears that these metals may not significantly impact the quality of recovered coagulants. Results of finished water quality testing at full-scale plants (Bishop et al. 1987; Magara, 1988a-g) indicate that trace metal contaminants are not carried over to the finished water, but are removed in the water treatment process. Further investigation into recovered coagulant impurity concentrations and the use of standard Water Chemicals Codex (National Research Council 1982) procedures for determination of these values is needed and will provide a much stronger basis for examination of recovered coagulant quality.

Soluble organic carbon (SOC) concentrations and organic priority pollutants contained in recovered coagulants were investigated by Saunders (1989). In

general, SOC concentrations were approximately equivalent to recovered aluminum concentrations and would contribute substantially to the SOC that must be removed in the water treatment process. Furthermore, SOC represents an increased potential for THM formation, but requires further investigation to determine possible impacts on finished water quality. Bishop et al. (1987) presented an investigation of THM formation potential (THMFP) during reuse of recovered coagulants. A slight increase in finished water THMFP during use of recovered coagulant was experienced, but further research is required in this area to properly address this problem. Organic priority pollutants in raw alum sludges, recovered coagulants and commercial coagulants were investigated by Saunders (1989). Although six organic compounds were detected in the raw alum sludges, no organic priority pollutants were detected in resulting recovered coagulants.

UTILITY OF RECOVERED COAGULANTS

Utility of recovered coagulants has been reported by numerous investigators at the laboratory-, pilot- and full-scale levels, however, full-scale operation provides the most reliable information on product utility. The Athens, TN facility typically applies recovered iron coagulants at doses equivalent to commercial coagulant application rates (Camp 1989). Recovered coagulant is added as the sole coagulant and has been found to produce finished water equivalent to that during use of commercial coagulants. During periods of poor raw water quality, commercial and recovered coagulants have been added simultaneously to ensure acceptable finished water quality. Although finished water quality appears to be equivalent during use of recovered and commercial coagulants, an increase in the lime dose for pH and alkalinity adjustment is required due to excess free acid associated with the recovered coagulants. Increased chlorine application rates have also been required for oxidation and removal of reduced compounds (e.g., manganese and ferrous iron). Referring to plant operations, an increased carryover of solids from sedimentation basins to filtration systems may be expected. This results in an increased solids loading on the filters and a decrease in the maximum length of a filter run prior to backwash during use of recovered coagulants. However, finished water turbidity at the plant is not impacted due to the use of recovered coagulant.

Full-scale operations in Japan use simultaneous application of recovered and commercial coagulants. Recovered and commercial coagulants are typically applied at near equal mass-application rates. Finished water quality reports indicate an acceptable finished-water quality resulting from this treatment scheme.

TREATMENT OF RESIDUAL SLUDGE SOLIDS

One of the main objectives of the coagulant recovery process is the reduction of sludge mass and volume for ultimate disposal through the dissolution of the voluminous aluminum and iron hydroxide sludge solids. Acidification of water treatment plant sludges results in the dissolution of sludge solids in the form of $Al(OH)_3 \cdot 3H_2O$ and $Fe(OH)_3 \cdot 3H_2O$ to numerous soluble monomers and polymers. This results in a dramatic decrease in the mass of residual sludge solids in proportion to the mass of coagulant-metal hydroxide dissolved. Therefore, sludges containing higher amounts of coagulant metal hydroxide solids should experience a higher degree of solids reduction with the use of coagulant recovery. Bishop et al. (1987) reported 53 and 42 percent reduction in residual sludge solids following acidification of alum sludge solids during full-scale testing at Durham, NC. Other small-scale investigations indicate that a solids reduction ranging from 9 percent (Cornwell and Susan 1979) to 86 percent (Cornwell and Susan 1979) is possible following acidification, but the reduction is highly dependent on sludge characteristics.

The reduction in the volume of residual sludge solids is dependent on the degree of solids separation obtainable following acidification, e.g., the volumetric recovery of clarified liquid following acidification. Gravity settling of residual sludge solids has been employed by numerous investigators for solids separation, while other have applied acidified sludge solids directly to vacuum dewatering beds and collected the bed filtrate as recovered coagulant (Camp 1989; American Water Works Service Company 1987). The volumetric reduction in residual sludge solids is highly dependent on sludge characteristics as indicated by broad variations, ranging from 36 percent (White and White 1985) to 92.5 percent (Webster 1969) of the initial sludge volume.

Numerous methods have been employed for treatment of residual sludge solids following acidic coagulant recovery. Direct application of acidified sludge solids to vacuum-assisted drying beds and filter press dewatering units have been investigated, while conditioning of acidified sludge solids with polymer and lime prior to dewatering on vacuum-assisted drying beds and filter press dewatering units have been effectively utilized at the full-scale operational level (Camp 1989; Magara 1988a-g). Therefore, the effectiveness of the coagulant recovery process for the reduction of sludge mass and volume for disposal has been demonstrated at the full-scale level. However, the methods employed for conditioning and dewatering of residual sludge solids and the degree of sludge solids volume and mass reductions requires further investigation for optimization of chemical additions and sizing of dewatering units.

ECONOMIC ANALYSIS OF COAGULANT RECOVERY

An extensive economic analysis of coagulant recovery chemical costs was conducted for the Athens, TN facility when recovered coagulant was used for treatment of 35 percent of the total potable water produced. The economic analysis of chemical costs included sulfuric acid and polymer used in the production of recovered coagulant, savings associated with decreased use of commercial coagulant and increased costs for lime and chlorine in the water treatment system during use of the recovered coagulants. The results indicate that chemical costs at the Athens facility were equivalent during periods of commercial and recovered coagulant use. Total chemical costs during use of recovered coagulants were $\$10.58/m^3$ ($\$40.03/10^6$ gal), while chemical costs during the use of commercial coagulants were $\$10.65/m^3$ ($\$40.30/10^6$ gal).

Other investigators reported the economic implications for the use of full-scale coagulant recovery. Operating and maintenance costs reported by three independent investigators were normalized to raw water flow and mass production of solids and compared. Operating and maintenance costs for proposed coagulant recovery systems at the Athens, TN (White and Company, 1983) and Spartanburg, SC (Jordan Jones and Goulding 1988) facilities were estimated to be $\$14.40/10^3$ m^3 ($\$54.50/10^6$ gal) and $\$3.30/10^3$ m^3 ($\$12.40/10^6$ gal), respectively. Operating and maintenance costs normalized to dry solids

production were similar for the AWWSC (1987) and Spartanburg, SC (Jordan, Jones and Goulding 1988) analyses at $0.45/kg ($0.21/lb) and $0.40/kg ($0.18/lb), respectively. Capital costs reported by the three independent investigators were normalized to daily raw water flow and daily mass production of solids and compared. The projected capital costs at Athens, TN and Spartanburg, SC were similar at approximately $23,800/10^3 m^3 ($90,000/10^6 gal). Capital costs normalized to dry solids production for AWWSC and Spartanburg, SC investigations were $1555/kg ($707/lb) and $2818/kg ($1281/lb), respectively. In general, the capital cost projections of the three investigators were comparable.

In all cases above, costs were compared between coagulant recovery systems and other solids handling systems. The results indicate that as solids production increases, the relative economic effectiveness of coagulant recovery increases. At both the Spartanburg, SC and Athens, TN facilities, the preliminary economic analyses indicated that the costs of coagulant recovery systems were similar to or less than other solids handling systems investigated. The economic analysis by American Water Works Service Company (1987) indicated that coagulant recovery becomes economically advantageous as solids production increases. Furthermore, in areas where land area is limited, coagulant recovery systems provide an alternative solids dewatering facility requiring significantly less area than many other systems. Bishop et al. (1987) determined that the use of coagulant recovery at Durham, NC would result in a 2.7-fold decrease in required drying bed area. Based on land area alone, coagulant recovery may be a viable option for conditioning and dewatering of water treatment plant sludges.

A main factor influencing the cost effectiveness of coagulant recovery systems is the cost for disposal of residual sludge solids. As demonstrated at Spartanburg, SC where sludge is disposed of in an authority-owned-and-operated landfill, when disposal costs are relatively inexpensive, coagulant recovery may not be an economically attractive alternative. However, in densely populated areas, such as Japan where sludge disposal costs are high and land area is limited, coagulant recovery offers a costs effective alternative for the conditioning and disposal of water treatment plant sludges.

INTRODUCTION

Treatment of surface waters for potable uses has long been accomplished using inorganic coagulants for the removal of turbidity and other trace contaminants. Coagulation, sedimentation and filtration processes are typically used in the destabilization, agglomeration and subsequent removal of contaminants that pose threats to public health or aesthetic quality of potable waters.

Metal salts, such as aluminum sulfate [$Al_2(SO_4)_3 \cdot 14.3H_2O$], aluminum chloride [$AlCl_3$], polyaluminum chloride [$(AlCl_3)_N$], ferric sulfate [$Fe_2(SO_4)_3 \cdot 3H_2O$], and ferric chloride [$FeCl_3$] are frequently used as coagulants. Inorganic coagulants destabilize colloidal dispersions to allow for agglomeration of colloidal particles and removal of inorganic and organic contaminants through adsorption, charge neutralization, complexation, precipitate enmeshment and polymer bridging mechanisms. The result of the coagulation process is the removal of a majority of contaminants from source waters to produce a water suitable for drinking and the production of a sludge containing metal hydroxides and oxides and organic and inorganic contaminants removed in the coagulation process.

COAGULATION PRACTICES IN WATER TREATMENT

Coagulation provides a means for removal of inorganic and organic contaminants in dissolved and suspended forms from source waters. Inorganic coagulants are typically used to facilitate destabilization of raw waters, enhancing agglomeration and removal of particulate matter. Naturally occurring humic substances and other organic constituents are removed primarily by the formation of metal complexes and adsorption onto the surface of the flocculated particles. Enmeshment in a precipitate (sweep coagulation) typically occurs when coagulant doses are sufficiently high to cause a rapid precipitation of a metal hydroxide which acts to enmesh many particles, enhance collisions with other particles and increase the surface area available for adsorption of organics. In general, a combination of particle destabilization and enmeshment can be considered to be the primary mechanism of coagulation (Weber 1980).

Chemistry of Coagulants

Use of pC-pH diagrams proves to be a useful tool in describing the metal species in solution following addition of a coagulant to water. A pC-pH diagram for aluminum is presented in Figure 1 (Weber 1980). Numerous positively-charged [Al^{3+}, $Al(OH)^{2+}$, $Al(OH)_2^{1+}$, and $Al_8(OH)_{20}^{4+}$] and negatively-charged [$Al(OH)_4^{1-}$] aluminum species exist in equilibrium with solid aluminum hydroxide [$Al(OH)_3 \cdot 3H_2O$], which has a minimum solubility at a pH of about 6.2.

Since aluminum is a hydrolyzing metal ion, addition of, for example, aluminum sulfate to a water results in an immediate coordination of the aluminum ion with six water molecules [$Al(H_2O)_6^{3+}$]. Hydrolysis reactions proceed instantaneously with the formation of numerous charged monomers [e.g., $Al(OH)^{2+}$, $Al(OH)_2^{1+}$], followed by formation of polymers [e.g., $Al_8(OH)_{20}^{4+}$, $Al_{13}(OH)_{34}^{5+}$], both of which approach or reach the equilibrium state presented in Figure 1. The polymer used in the development of this diagram is not inclusive of all aluminum polymerization products, but is to represent a typical polymer present during polymerization reactions, and is indicative of typical precursors of aluminum-hydroxide precipitates presented in the diagram.

The existence of various charged monomers and polymers is a function of pH and aluminum concentration and can be predicted based on a pC-pH diagram. Aluminum concentrations are expressed in Figure 1 in moles/L as Al, where one mole of aluminum is equivalent to 27 g of Al. At a pH near 6.2, the minimum solubility of aluminum, only about 10^{-7} moles/L (2.7 μg/L) of Al remains in soluble form, existing as numerous charged species as described earlier. As the pH of the solution varies from the point of minimum solubility, the soluble aluminum concentration increases as presented in Figure 1. At a pH value of 4.0, for example, numerous positively-charged aluminum species, such as $Al(OH)_2^+$, $Al(OH)^{2+}$, Al^{3+} and positively-charged polymerization products can be present at concentrations up to about 10^{-2} moles/L (270 mg/L) as Al. At a pH value of 10, the maximum soluble aluminum concentration is approximately 10^{-3} moles/L (27 mg/L) as Al. $Al(OH)_4^-$ is the predominant species in the alkaline pH range, although other negatively-charged species exist at significantly lower concentrations.

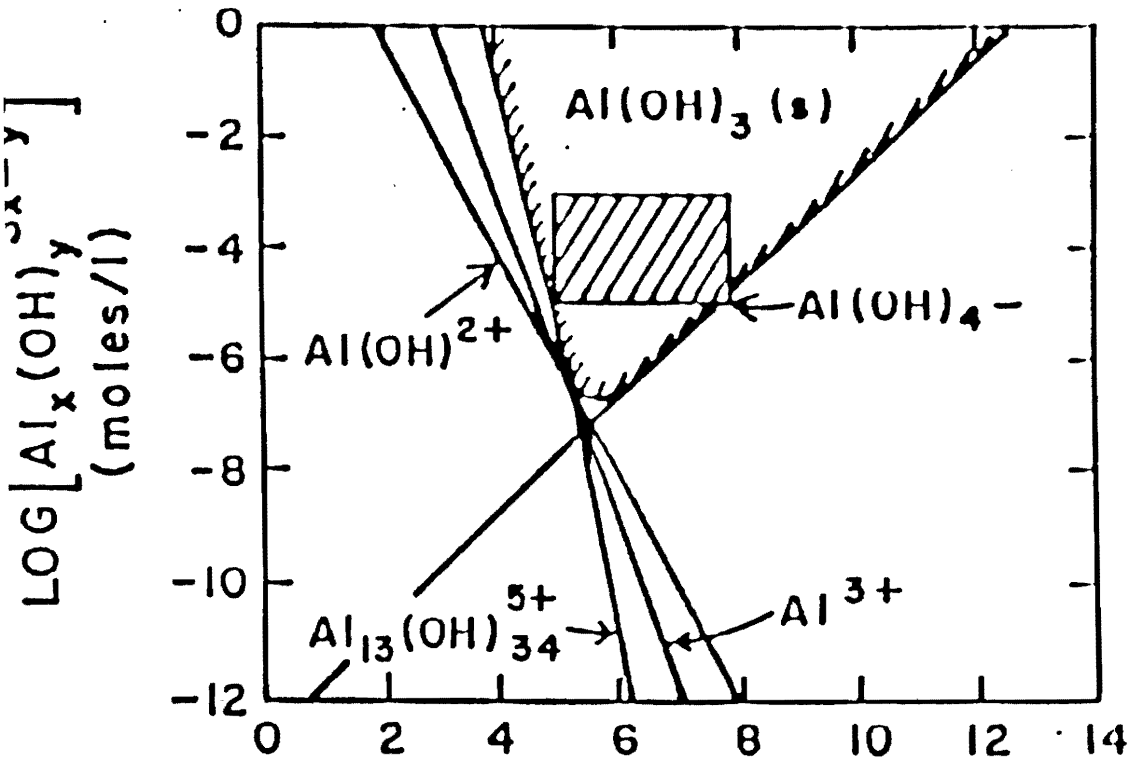

FIGURE 1. A pC-pH Diagram for Aluminum (Al-III) Species in Equilibrium with Solid Aluminum Hydroxide (Weber 1972)

Iron is also a hydrolyzing metal having a chemistry similar to that of aluminum. As presented in Figure 2, the pC-pH diagram for iron has many of the same characteristics as that for aluminum. Hydrolysis reactions cause the formation of monomers and polymers in equilibrium with solid iron hydroxide precipitates, which have a minimum solubility at a pH of about 8. At this point of minimum solubility, about 10^{-11} moles/L (0.558 ng/L) of Fe is soluble as numerous charged monomers [$Fe(OH)^{2+}$, $Fe(OH)_2^{1+}$, $Fe(OH)_4^{1-}$], dimers [$Fe_2(OH)_2^{4+}$] and polymerization products. As the pH deviates from the point of minimum solubility, the soluble iron concentration increases dramatically. For example, at a pH value of 10, 10^{-9} moles/L (55.8 ng/L) of Fe is soluble in the predominant form of $Fe(OH)_4^{1-}$. As the pH decreases, a more complex population of iron species is present. At a pH value of 4.0, approximately 10^{-6} mole/L (55.8 μg/L) iron is soluble in the form of $Fe(OH)^{2+}$, $Fe(OH)_2^{1+}$, $Fe_2(OH)_2^{4+}$ and Fe^{3+}.

A comparison of iron and aluminum equilibria and solubility shows that aluminum is significantly more soluble in the neutral pH range as compared to iron. For example, at a pH value of 4.0, approximately 270 mg/L of aluminum can be present in solution compared to 44.8 μg/L of soluble iron at the same pH. Furthermore, at the point of minimum solubility for each metal, 2.7 μg/L and 0.558 ng/L of aluminum and iron, respectively, can be present in solution, a difference of more than three orders of magnitude. The pH of minimum solubility of aluminum is about 6.2 compared to approximately 8.0 for iron. In general, iron is a stronger acid and much less soluble than aluminum.

Coagulant Application in Water Treatment

To minimize conflicts, it is advantageous to use common terminology for commercial aluminum coagulants. The term "alum" will be used herein to refer to commercial aluminum sulfate [$Al_2(SO_4)_3 \cdot 14.3H_2O$] coagulants, where 1 mole of alum is equivalent to 2 moles of Al or 599.4 g of alum is equivalent to 54 g of Al. This corresponds to a mass ratio of 0.0901 g Al/g alum.

Investigations of coagulation practices used in the industry and reported in the literature have led to the development of coagulation diagrams showing the predominant areas of coagulation with respect to a pC-pH diagram. For

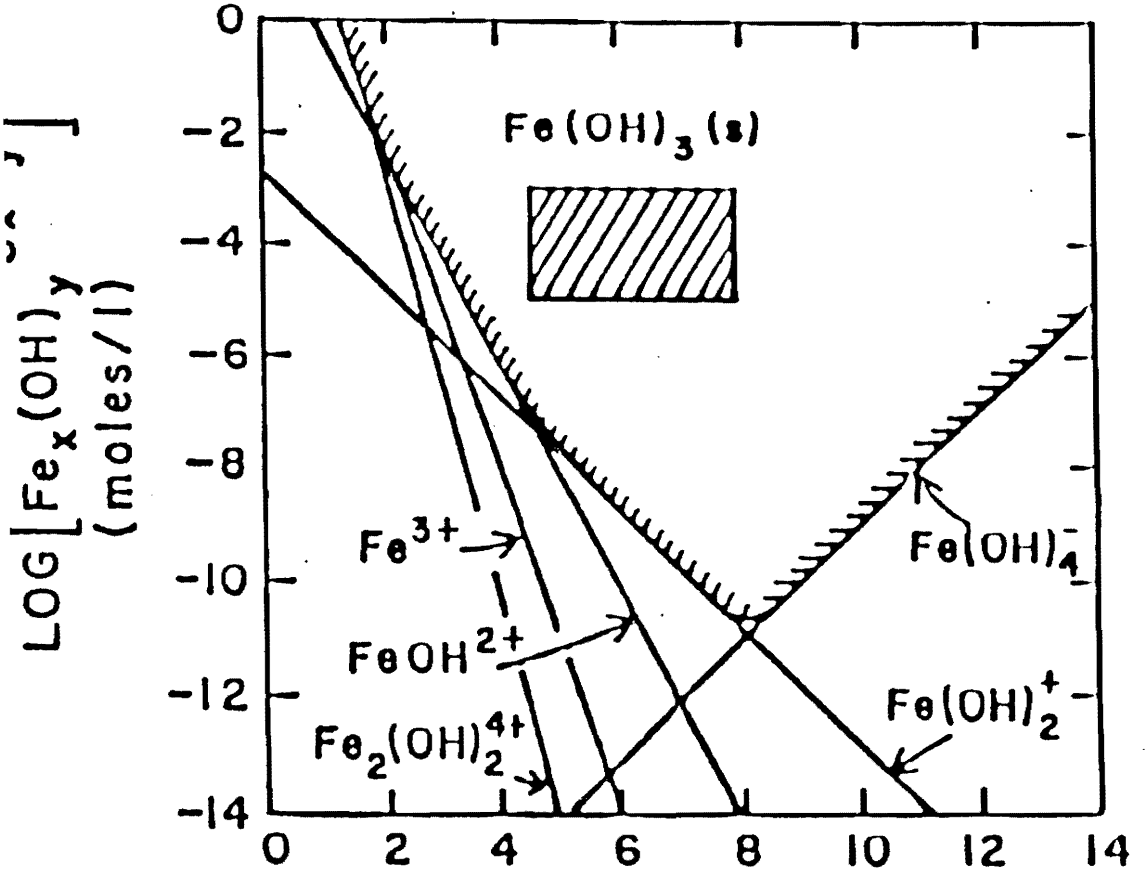

FIGURE 2. A pC-pH Diagram for Iron (Fe-III) Species in Equilibrium with Solid Iron Hydroxide (Weber 1972)

example, Edwards and Amirtharajah (1985) developed an alum coagulation diagram, as presented in Figure 3, by superimposing pertinent literature data onto a pC-pH diagram for aluminum hydroxide. Based on the data collected, predominance areas were established for sweep coagulation and particle destabilization.

As indicated in Figure 3, typical alum doses range from about 0.3 mg/L to greater than 150 mg/L, corresponding to aluminum doses of about 0.027 mg/L as Al to greater than 13.5 mg/L as Al. Although pH dependent, sweep coagulation generally occurs at alum doses above about 15 mg/L, or 1.35 mg/L as Al, with a combination of sweep coagulation and charge neutralization occurring at substantially lower doses.

An iron coagulation diagram was developed in much the same way as was that for aluminum. Johnson and Amirtharajah 1983 introduced the predominance areas for iron coagulation superimposed on the iron pC-pH diagram, as presented in Figure 4. In general the dose of ferric chloride ($FeCl_3 \cdot 6H_2O$) and was comparable to that of alum, in the range of 1 to more than 100 mg/L as $FeCl_3$ $6H_2O$ equivalent to 0.21 to 21 mg/L as Fe. Comparison to the aluminum coagulation diagram indicates that iron is an effective coagulant in a much broader pH range, while the mass application is similar.

CONTROL OF COMMERCIAL COAGULANT QUALITY

The American Water Works Association has developed standards for many of the chemicals used in the water treatment industry. The current standard for aluminum sulfate (AWWA 1982) was approved by the American National Standards Institute, Inc., in August of 1982 and specifies that

> "The aluminum sulfate supplied under this standard shall contain no soluble mineral or organic substances in quantities capable of producing deleterious or injurious effects on public health or water quality."

Iron is the only impurity in the commercial products which is specifically controlled under the current AWWA standard. In dry aluminum sulfate the iron content should not exceed 0.75 percent (as Fe_2O_3) based on 17 percent Al_2O_3; or 0.35 percent (as Fe_2O_3) in liquid alum based on 8.0 percent as Al_2O_3.

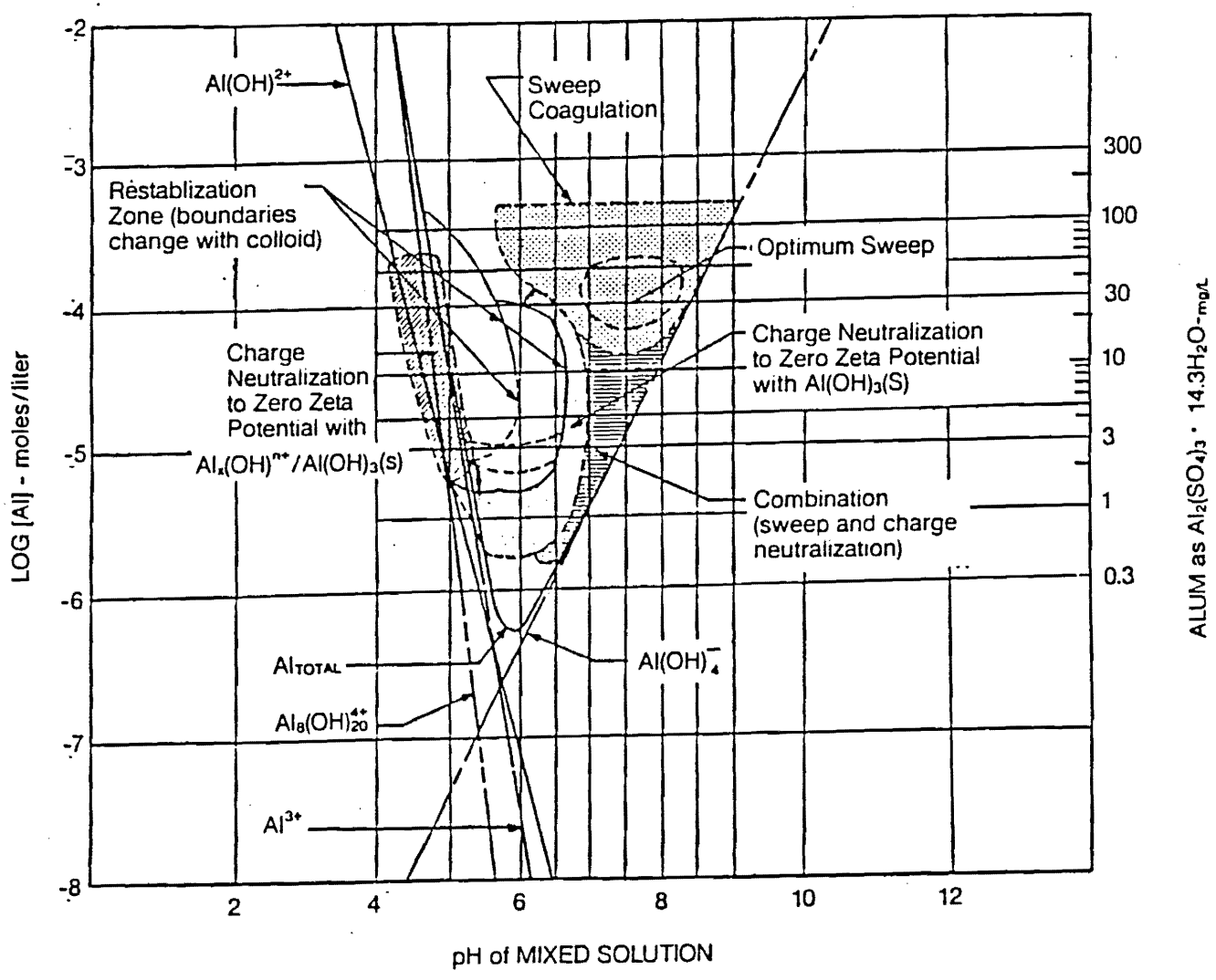

FIGURE 3. Alum Coagulation Diagram (Edwards and Amirtharajah 1985)

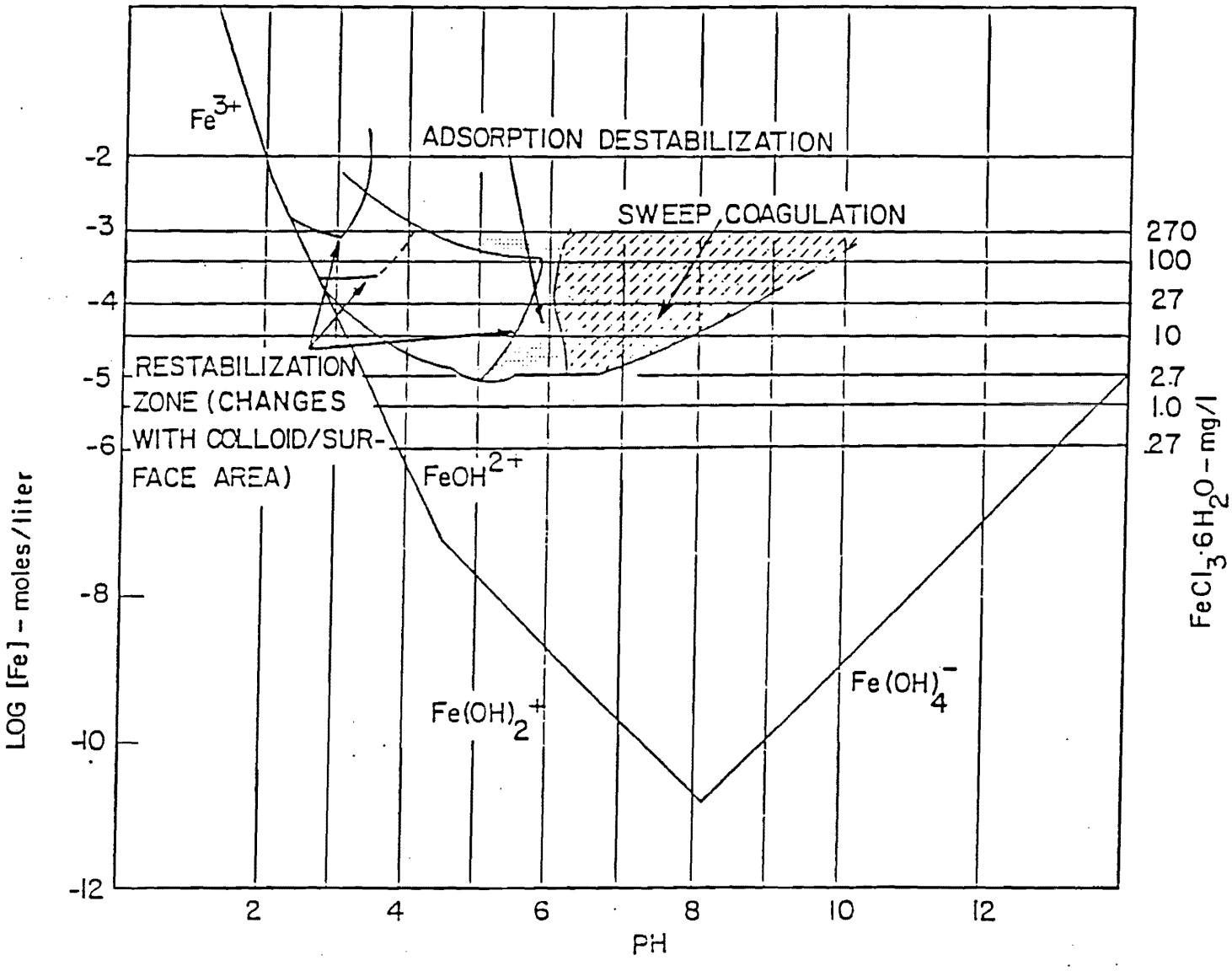

FIGURE 4. Iron Coagulation Diagram (Johnson and Amirtharajah 1983)

More specific guidelines have been established by the Water Chemicals Codex (National Research Council 1982) which provides criteria for purity requirements for chemicals used in the water treatment industry based on use patterns of the additive and contaminant toxicity. It should be stressed that the Water Chemicals Codex (National Research Council 1982) is not currently used for regulatory purposes, but is used to provide a guideline for product quality. Materials which are regenerated or reclaimed, such as recovered coagulants, are also covered under the provisions of the Water Chemicals Codex (National Research Council 1982).

A list of impurities to be considered was developed based on maximum contaminant levels, MCLs, as specified by the National Interim Drinking Water Act of 1974, as well as other contaminants commonly found in water treatment chemicals. Recommended maximum impurity contents (RMIC) for each impurity were calculated using MCL values for drinking water or a suggested no-adverse-response level, SNARL, based on known genotoxicity, threshold limit values, average body weight, intake and exposure to the contaminant. Recommended maximum impurity contents were also calculated using a safety factor (SF) of 10 and a maximum dosage (MD). The maximum dosage for a water treatment chemical was based upon maximum dosages found to be representative of coagulant doses used in the industry or at a particular plant. This value should be increased if maximum doses at a particular site exceed that recommended, with RMIC values being adjusted accordingly. A maximum dose of 150 mg/L was recommended for alum used as a coagulant in water treatment. Therefore, calculation of an RMIC value would be as follows:

$$\text{RMIC} = \frac{\text{MCL}}{\text{MD} \times \text{SF}} \qquad (1)$$

For example, an RMIC calculation for an impurity with an MCL of 0.002 mg/L, a safety factor of 10 and a maximum dose of 150 mg/L of alum would be as follows:

$$\text{RMIC} = \frac{0.002 \text{ mg/L} \times 10^6 \text{mg/kg}}{150 \text{ mg/L} \times 10} = 1.33 \text{ mg/kg alum} \qquad (2)$$

Concentrations are also expressed as mg/kg aluminum, as it will be used in the case of recovered coagulants for which aluminum concentrations are typically

expressed. The RMIC value calculated above is equivalent to about 15 mg/kg Al. RMIC values for aluminum sulfate presented in Table 1 represent recommended maximum impurity contents as specified by the Water Chemicals Codex (National Research Council 1982) based on a maximum alum dose of 150 mg/L and are intended for use as a guideline for the industry. RMIC values for arsenic, chromium, lead and silver are 330 mg/kg Al (30 mg/kg alum), while values for cadmium and selenium are 77 mg/kg Al (7 mg/kg alum). Mercury has the lowest RMIC value at 11 mg/kg Al or 1 mg/kg alum.

TABLE 1

Recommended Maximum Impurity Content (RMIC) Values for Aluminum Sulfate Coagulants According to Water Chemicals Codex (National Research Council 1982)

	RMIC	
Impurity	mg/kg alum	mg/kg Al
Arsenic	30	330
Cadmium	7	77
Chromium	30	330
Lead	30	330
Mercury	1	11
Selenium	7	77
Silver	30	330

Specific analytical techniques are described in the Water Chemicals Codex (National Research Council 1982) for determination of impurity contents. The analytical procedures for aluminum sulfate required making a 1-L solution of alum at a concentration of 150 mg/L. The pH of the solution should then be adjusted to 6 using 1 N NaOH and the suspension allowed to stand for one hour. Following membrane filtration, nitric acid digestion of the filtrate should be employed and the sample tested for arsenic, cadmium, chromium, lead, mercury, selenium, and silver. Values obtained should be normalized to the 150 mg/L of alum used initially (i.e., mg impurity/kg alum) and compared to calculated RMIC values presented in Table 1.

Recommended maximum impurity contents have also been established for other chemicals used in the water treatment industry. RMIC values for ferric

sulfate will be presented herein, as they will be employed later. A maximum dosage for ferric sulfate [$Fe_2(SO_4)_3 \cdot 5H_2O$] for 100 mg/L was established by the Water Chemicals Codex (National Research Council 1982) based on use patterns in the industry. The appropriate RMIC values are presented in Table 2. RMIC values for arsenic, chromium, lead and silver are 152 mg/kg of Fe (50 mg/kg ferric sulfate), while values for cadmium and selenium are 30 mg/kg of Fe (10 mg/kg ferric sulfate). Mercury has the lowest RMIC value at 6 mg/kg of Fe (2 mg/kg ferric sulfate).

TABLE 2

Recommended Maximum Impurity Content (RMIC) Values for Ferric Sulfate Coagulants According to Water Chemicals Codex (National Research Council 1982)

	RMIC	
Impurity	mg/kg $Fe_2(SO_4)_3 \cdot 5H_2O$	mg/kg Fe
Arsenic	50	330
Cadmium	10	30
Chromium	50	152
Lead	50	152
Mercury	2	6
Selenium	10	30
Silver	50	152

QUALITY OF COMMERCIAL COAGULANTS

Bauxite ores and clays are typically used as aluminum sources for the production of commercial-grade alum. Aluminum is present in these raw materials in the form of various aluminum oxides, $Al_2O_3 \cdot nH_2O$, requiring digestion with sulfuric acid at a temperature of about 100-120°C for dissolution of aluminum. Other acid soluble components of the raw materials are also dissolved, appearing as contaminants in the finished product. Commercial-grade alum is typically marketed in a liquid form at 8.25 percent Al_2O_3, which is equivalent to approximately 56 g/L of aluminum in a solution with a specific gravity of 1.3. Iron-free alum, which is typically certified to contain iron at <50 ppm as Fe_2O_3, is produced from Bayer Process precipitator scale and is of much

higher purity relative to trace contaminants than typical commercial-grade alum used in water treatment. The Bayer Process is the process by which aluminum is extracted from bauxite ores using sodium hydroxide. Iron-free alum coagulants produced from this process typically cost twice as much as standard commercial alum and are infrequently used in potable-water treatment.

Commercial alum has been investigated by Cornwell and Westerhoff (1982), Bishop et al. (1987) and Saunders (1989) for determination of aluminum and contaminant concentrations, as presented in Table 3. Aluminum concentrations ranged from 55.05 g/L in the Atlanta #2 alum sample to 63.0 g/L in the Durham, N. C. alum samples, corresponding closely to the manufacturing specification of 56 g/L. Iron was found to be the most predominant contaminant in all of the coagulants, ranging from 850 mg/L to 2080 mg/L in the Durham, Tampa and Atlanta samples. The iron-free alum contained only about 10.1 mg/L due to the higher quality of the materials used to produce iron-free alum. Chromium was the next highest constituent in commercial alum used in water treatment plants at concentrations from 0.9 mg/L to 77.5 mg/L. Manganese concentrations ranged from 1.7 mg/L to 20 mg/L, while zinc was detected at 0.2 mg/L to 8.5 mg/L. Lead and copper were also detected at concentrations ranging up to 10 mg/L and 5.5 mg/L, respectively. Although other heavy metals were detected in commercial alum samples, they were at significantly lower concentrations.

Impurity concentrations were normalized to aluminum (i.e., mg/kg Al) for comparison to purity requirements in the Water Chemicals Codex (National Research Council 1982) and are presented in Table 4. It should be noted that these concentrations were determined based on acid-digestion procedures set forth in Standard Methods (APHA 1985) for determination of total metal concentration in an alum product, not the procedures recommended by the Water Chemicals Codex (National Research Council 1982) which provides for removal of contaminants in conjunction with coagulation at pH 6. These normalized impurity concentrations, however, will be compared with RMIC values from the Water Chemicals Codex (National Research Council 1982) recognizing this unfavorable bias. Furthermore, if an impurity concentration is less than the RMIC value then this is a strong indication of the acceptable quality of the coagulant, due to the conservative method of analysis.

TABLE 3

Metal Content in Commercial Alum Coagulants (Bishop et al. 1987, Cornwell and Westerhoff 1982 and Saunders 1989)

	Metal Concentration, mg/L				
	Commercial Alum				
Component	Durham[1]	Tampa[2]	Atlanta[3]	Atlanta[3]	Commercial Iron-Free Alum[3]
Aluminum	63000	50000	60300	55050	56000
Arsenic	3.0	--	--	--	--
Barium	0.5	0.1	*	*	--
Cadmium	*	0.4-0.7	0.025	0.3	0.3
Calcium	6.3	--	--	--	--
Chromium	9.5	2.6-22	77.5	40	0.9
Cobalt	--	1.7-3.0	--	--	--
Copper	0.1	0.8-1.5	5.5	2.8	--
Iron	1160	850	1845	2080	10.1
Lead	1.5	9-10	6.6	4.06	5.6
Magnesium	12.5	5.5-33	--	--	--
Manganese	1.7	1.8-7.0	20	20	--
Mercury	0.001	--	0.0048	0.0032	--
Nickel	0.1	--	43.5	43.5	--
Potassium	5.6	--	--	--	--
Selenium	*	--	--	--	--
Silicon	14.2	--	--	--	--
Silver	0.4	0.1-0.6	0.25	0.2	0.15
Sodium	57	--	--	--	--
Tin	--	--	155	155	2.5
Zinc	1.1	0.2-1.7	8.5	8.5	1.0

*Below Detection Limit
[1]Bishop, et al. 1987
[2]Cornwell and Westerhoff 1982
[3]Saunders 1989

TABLE 4

Metal Content Normalized to Aluminum Content for Commercial Alum Coagulants and a Commercial Iron-Free Alum (Bishop, et al. 1987, Cornwell and Westerhoff 1982 and Saunders 1989)

Component	RMIC[1] mg/kg Al	Impurity Content, mg/kg Al				
		Commercial Alum				Commercial Iron-Free[4]
		Durham[2]	Tampa[3]	Atlanta[4]	Atlanta[4]	
Arsenic	330	47.6	--	--	--	--
Barium	*	7.9	2.0	--	--	--
Cadmium	77	--	8-14	0.41	5.4	5.4
Calcium	*	100	--	--	--	--
Chromium	330	150.8	52-440	1285	726.6	16.1
Cobalt	*	--	34-60	--	--	--
Copper	*	1.6	16-30	91.2	50.9	--
Iron	*	18413	17000	30600	37780	180
Lead	330	23.8	180-200	298.5	181.6	100
Magnesium	*	198	110-660	--	--	--
Manganese	*	27.0	36-140	331.7	363.3	--
Mercury	11	0.02	--	0.08	0.06	--
Nickel	*	1.6	--	723.1	844.7	--
Potassium	*	88.9	--	--	--	--
Selenium	77	--	--	--	--	--
Silicon	*	225.4	--	--	--	--
Silver	330	6.3	2-12	4.15	3.63	2.7
Sodium	*	905	--	--	--	--
Tin	*	--	--	2570	2815.6	44.6
Zinc	*	17.5	4-34	141	154.4	17.9

*Not specified by Water Chemicals Codex (National Research Council 1982)
[1]RMIC value based on 150 mg/L as alum (13.5 mg Al/L) and a safety factor of 10 (National Research Council 1982)
[2]Bishop, et al. 1987
[3]Cornwell and Westerhoff 1982
[4]Saunders 1989

Chromium concentrations were high in all commercial alum samples, with the concentration exceeding the RMIC of 330 mg/kg Al in the Tampa and both Atlanta samples. Lead concentrations were also notably high, but did not exceed the RMIC of 330 mg/kg Al. Tin and nickel concentrations in the Atlanta samples were >2500 mg/kg Al and >700 mg/kg Al, respectively, although no RMIC has been established. Iron concentrations were also high in the samples, ranging from 17000 to 38000 mg/kg Al. Data for the iron-free commercial alum indicate lower impurity concentrations as compared to the other commercial alum studied. Normalized iron concentrations were significantly lower at 180 mg/kg Al compared to greater than 17000 mg/kg in the other samples.

SLUDGE PRODUCTION AND CHARACTERISTICS

Metal hydroxide sludges are produced in conjunction with coagulation of colloids and contaminants contained in raw waters used in the treatment of potable drinking water. Alum sludges have been described as "non-Newtonian, bulky, gelatinous substances composed primarily of metal hydroxides and organic and inorganic particles, such as clay, sand, color, colloids, microorganisms and other matter removed from the raw water" (Albrecht 1972). Numerous investigators have presented the physical and chemical characteristics of both aluminum and iron sludges, the results of which are presented herein.

Aluminum Hydroxide Water Treatment Plant Sludges

Chen et al. (1975) used a pC-pH diagram for aluminum hydroxide to investigate aluminum species present based on basic thermodynamic properties of aluminum. It was concluded that the typical operational pH and concentration range employed in water treatment would fall within the region where $Al(OH)_3 \cdot 3H_2O$ is the predominant form of aluminum present at equilibrium. For example, in a water treatment plant treating water from a run-of-the-river source, a typical alum dose would be about 30 mg/L (2.7 mg/L as Al) and the majority of the aluminum in the sludge would be expected to be present in the form of aluminum hydroxide [$Al(OH)_3 \cdot 3H_2O$]. More recent investigations into alum coagulation by Edwards and Amirtharajah (1985) have shown typical areas for coagulation to fall within the area where aluminum hydroxide would be predominant, as shown

in Figure 3. From these data and others, it is reasonable to indicate that $Al(OH)_3 \cdot 3H_2O$ is the predominant form of aluminum in an alum sludge produced at typical pH and concentration ranges used in the water treatment industry.

Aluminum hydroxide precipitates typically contain high amounts of bound water, making alum sludge a voluminous suspension, with low solids concentrations and poor dewatering properties. Solids concentration, pH and aluminum concentration of aluminum hydroxide sludges, as reported by many sources, are presented in Table 5. Solids concentrations, settling velocity and specific resistance are presented in Table 6. Total solids ranged from about 4.5 to 121 g/L, while aluminum concentration ranged from 300 to about 21000 mg/L. The pH of the sludge suspensions ranged from a value of 6.5 to about 7.7. Initial settling velocity was about 0.1 cm/sec where reported and specific resistance ranged from about 1 to 80 Tm/kg.

Sludge data collected from four water treatment plants in Atlanta, Georgia are presented in Table 7 (Saunders 1989). Although these data do not necessarily represent the suspended solids concentrations encountered at the site due to short-term gravity thickening of the samples following collection, the data indicate the varying nature of the alum sludges. Suspended solids on the Chattahoochee sludge samples ranged from 50.7 to 179.4 g/L, while the pH ranged from 5.8 to 7.3. The Hemphill sludges varied significantly in solids concentrations from 8.67 to 120.3 g/L while the pH ranged from 6.4 to 7.7. The Candler and Quarles samples had suspended solids concentrations of 10.85 g/L and about 100 g/L, respectively, with pH values ranging from 5.95 to 6.8. Total sludge aluminum content was measured on four sludge samples and varied from 831 mg/L as Al for the Candler sludge to 5167 mg/L as Al for the Quarles sludge. Capillary suction time (CST) is presented in Table 7 for the Atlanta sludges and provides an indication of the filterability of the sludge suspensions. The Hemphill and Candler sludges had CST values in the range of 20-60 sec, while the Chattahoochee and Quarles sludge samples were significantly more difficult to dewater with CST values ranging from 140 to 830 sec, due in part to higher suspended solids concentrations.

TABLE 5

Sludge Solids, pH and Aluminum Concentrations of Raw Alum Sludge Suspensions Reported by Numerous Investigators

Location	Solids			pH	Aluminum Concentration	Reference
	Total g/L	Suspended g/L	Volatile %		mg/L	
Durham, NC	--	24	--	7.13	--	Bishop et al. 1987
Durham, NC	--	33.5	--	6.64	--	Bishop et al. 1987
British Water Works	--	40-60	--	6.5-7.3	--	Isaac and Vahidi 1961
British Water Works	39.8	--	43.5	--	21000	Isaac and Vahidi 1961
Erie County, NY						
Coagulation Basin	--	40-60	--	--	--	Gruninger 1975
Filter Backwash	--	10-30	--	--	--	Gruninger 1975
Virginia						
Radford	29.8	--	31.7	6.7	3480	Chen et al. 1976
Timberville	13.5	--	22.1	7.1	1175	Chen et al. 1976
Harrisonburg	21.0	--	26.2	7.2	1620	Chen et al. 1976
Blacksburg	30.8	--	30.0	6.85	1383	Chen et al. 1976
Tampa	22.0	--	--	7.3	1700	Pryzbla et al. 1979
Sharon #1	19.0	--	--	7.4	1700	Pryzbla et al. 1979
Sharon #2	36.0	--	--	7.7	2200	Pryzbla et al. 1979
Milwaukee	8.05	7.75	--	7.0	--	Albrecht, 1972
Indianapolis	17.0	--	--	--	2400	Cornwell and Susan 1979
Concord	17.0	--	--	--	2400	Cornwell and Susan 1979
Tampa	16.0	--	--	--	3500	Cornwell and Susan 1979
Moline	17.0	--	--	--	295	Cornwell and Susan 1979
Washington	121.0	--	--	--	3750	Cornwell and Susan 1979
Atlanta	4.75	--	--	6.7	5200	Holmes 1982
Atlanta	11.20	--	--	6.5	6000	Holmes 1982
Atlanta	4.5	--	--	6.8	3000	Holmes 1982

TABLE 6

Sludge Solids, Initial Settling Velocity and Specific Resistance of Raw Sludge Suspensions Reported by Numerous Investigators

Location	Solids		Initial Settling Velocity	Specific Resistance	Reference
	Total g/L	Volatile %	cm/s	Tm/kg	
British Water Works	39.8	43.5	--	75.5	Isaac and Vahidi 1961
Virginia					
Radford	29.8	31.7	0.060	1.65	King et al. 1975
Timberville	13.5	22.1	0.074	5.4	King et al. 1975
Harrisonburg	21.0	26.2	0.095	3.6	King et al. 1975
Blacksburg	30.8	30.0	0.150	4.1	King et al. 1975

TABLE 7

Raw Alum Sludge Characteristics of Samples Collected from Four Water Treatment Plants in Metropolitan Atlanta, GA (Saunders 1989)

Sample	Suspended Solids g/L	pH	CST (10 mm) sec	Total Aluminum Concentration mg/L
Chattahoochee				
C-07-19-S	179.4	6.45	603	4904
C-07-22-S	104.19	6.42	405	-
C-08-01a-S	142.9	5.79	289	-
C-08-01b-S	77.6	5.88	139	-
C-08-21-S	171.7	6.1	678	-
C-08-29-S	235.9	7.29	237	-
C-09-27-S	50.7	-	7*	-
Hemphill				
H-07-22-S	8.67	6.38	22	895
H-08-29-S	120.3	7.72	59	-
H-09-10d-S	23.5	7.2	23	-
Candler (DeKalb)				
D-08-01-S	10.85	5.95	61	831
Quarles (Cobb)				
Q-08-01-S	97.4	5.96	830	5167
Q-09-18-S	103.7	6.8	311*	-
QW-09-16-S	-	-	-	-

*CST (18 mm)

Saunders (1989) reported sludge metal content (mg/L) and normalized sludge metal content (mg/kg suspended solids), as presented in Table 8. Aluminum was the most predominant component in the sludge ranging from 831 to 5167 mg/L or 47068 to 76590 mg/kg SS. Iron was abundant in all sludge samples ranging from 490 to 4900 mg/L, and was on the same order of magnitude as aluminum. Normalized iron concentrations ranged from 39908 to 56517 mg/kg SS. Manganese was the next most predominant metal at concentrations of 39 to 249 mg/L or 1283 to 4614 mg/kg SS. Barium was high in two sludge samples at concentrations of 20 mg/L (205 mg/kg SS) and 25 mg/L (240 mg/kg SS), while lead was present in all sludge samples at concentrations ranging from 0.56 to 6.85 mg/L. Chromium was also notably high, ranging from 0.5 to 6.0 mg/L. Normalized chromium concentrations ranged from 46.1 to 108.9 mg/kg SS.

Iron Hydroxide Water Treatment Plant Sludges

As indicated earlier, the chemistry of iron coagulation is similar to that of aluminum. Because of this, much of the above information is generally applicable to iron sludges produced from the coagulation of raw waters. The iron coagulation diagram presented earlier in Figure 4 indicated that the predominant form of iron in an iron sludge can be represented by $Fe(OH)_3 \cdot 3H_2O$. Limited data exists on the characteristics of iron sludges due to the limited use of iron coagulants in the water treatment industry.

Examination of data presented in Table 9 for four iron sludges indicates sludge characteristics are highly variable. Total solids concentrations ranged from 20 to 52 g/L, while iron concentration ranged from 930 to 4100 mg/L. The pH of the iron sludge was higher than that for alum sludge, ranging from 7.4 to 8.5.

As is evident from data presented above on water treatment sludges, there is no typical range of values for the various characteristics of water treatment sludges. Solids content, coagulant metal concentrations, pH, filterability and thickening properties vary considerably based on chemical additions and plant operations. In addition, characteristics change with varying raw water quality and also seasonally at an individual plant.

TABLE 8

Total Metal Composition of Raw Sludges Collected from Four Water Treatment Plants in Metropolitan Atlanta, GA (Saunders 1989)

	C-07-22		H-07-22		D-08-01		Q-08-01	
Metal	mg/L	mg/kg SS	mg/L	mg/kg SS	mg/L	mg/kg SS	mg/L	mg/kg SS
Al	4,904	47,068	885	102,076	831	76,590	5,267	53,049
Ag*	0.024	0.23	0.004	0.46	.00275	0.25	0.014	0.14
Ba*	25	240	<20	-	<20	-	20	205
Cd*	0.011	0.106	.02	2.3	0.016	1.5	0.65	0.67
Cr*	5.5	52.8	0.944	108.9	0.5	46.1	6.0	61.1
Cu	4.15	39.8	0.54	62.3	0.415	38.3	4.925	50.6
Fe	4,158	39,908	490	56,517	518	47,742	4,900	50,308
Hg*	0.004	0.04	0.0017	0.20	0.0028	0.26	0.019	0.19
Mn	249	2,390	40	4,614	39	3,594	125	1,283
Ni	2.6	25.0	0.5	57.7	0.5	46.1	2.4	24.6
Pb*	6.1	58.6	1.45	167.2	0.56	51.6	6.85	70.3
Sn	10	92.17	<10	-	<10	-	10	102.7
Zn	13	124.8	1.6	184.5	1.0	92.2	9.4	96.5

*Metals for which Maximum Content Levels (MCL) are established for finished drinking water.

TABLE 9

Characteristics of Raw Iron Sludge Collected from Water Treatment Plants Using Commercial Iron Coagulants

	Kingsport[1]	Kingsport[2]	St. Louis County[2]	Unspecified
Solids				
Total	40,800 mg/L	20,000 mg/L (2.0%)	20,000 mg/L (2.0%)	4.4-5.2%
Volatile	6,895 mg/L	2,800 mg/L (14% TS)	1,000 mg/L (5% TS)	--
Suspended Solids				
Total	--	19,900 mg/L (1.99%)	19,800 mg/L (1.98%)	--
Volatile	--	2,800 mg/L (14.1% TSS)	1,000 mg/L (5.1% of TSS)	--
Iron Content	2,350 mg/L as Fe	4,120 mg/L as Fe (20.6% of TS)	930 mg/L as Fe (4.65% of TS)	--
pH	7.6	7.4	8.5	--
Specific Resistance, Tm/kg	19.5	--	--	--
Settling Velocity, cm/s	0.007	--	--	--

[1]King et al. 1975
[2]Pigeon 1976
[3]White and White 1984

DISPOSAL OF WATER TREATMENT PLANT SLUDGES

Disposal of water treatment plant sludges is regulated by the Resource Conservation and Recovery Act (RCRA), the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) and the Clean Water Act (CWA). Regulatory acts governing the disposal of water treatment plant sludges based on disposal technique are presented in Table 10 (Cornwell et al. 1987). In-stream standards for contaminants have been set which must be met where direct discharge is employed. Many municipal and industrial solid waste landfills currently accept water treatment plant sludges, but may require a minimum water content or passage of the EP toxicity test for hazardous wastes (Cornwell et al. 1987).

TABLE 10

Regulatory Acts Governing Water Treatment Plant Waste Disposal
(Cornwell et al. 1987)

Disposal Option	Applicable Regulations
Stream	NPDES (CWA) In-Stream Water Quality Criteria (CWA) Discharge Guidance Documents
Wastewater Plant	Pretreatment Standards (CWA)
Landfill	RCRA CERCLA State SW Requirements (RCRA) Low Level Radioactive Waste Requirements (State, NRC, DOT, EPA)
Land Application	Sludge Disposal Regulations (CWA) Low Level Radioactive Waste Requirements (State, NRC, DOT, EPA)

Note: CWA = Clean Water Act
RCRA = Resource Conservation and Recovery Act
CERCLA = Comprehensive Environmental Response, Compensation and Liability Act

Cornwell and Susan (1979) reported the results of a survey of 75 water treatment plants in the United States. The number of plants using a particular disposal technique and the percentage of total flow surveyed using each technique are presented in Table 11. Permanent lagoons appeared to be the most popular method of disposal with 43 percent of the plants surveyed (or 70 percent of the total flow surveyed) employing this technique. Sewer discharge was practiced by 27 plants surveyed or 27 percent of the total flow surveyed, while 20 plants (1 percent of the total flow) practiced direct discharge to receiving streams.

TABLE 11

Alum Sludge Disposal Methods Reported by 75 Respondents in a 1975 Survey of Water Treatment Plants (Cornwell and Susan 1979)

Sludge Disposal Method	Percentage of Total Flow Surveyed	Percentage of Plants Surveyed
lagoon	70	43
sewer	27	27
rivers and lakes	1	20
other	2	10

Due to the increasing regulations and costs of sludge disposal, some plants are looking towards more cost effective sludge treatment and disposal techniques. Many options are available for thickening, conditioning, dewatering and ultimate disposal of water treatment plant sludges. As presented in Figure 5 (Cornwell et al. 1987), sludge from the underflow of the clarifier can be thickened prior to conditioning and dewatering. Lime conditioning, polymer conditioning or coagulant recovery can be employed prior to using lagoons, sand beds, centrifugation, pressure filtration, vacuum filtration or belt filter presses for final dewatering. Finally, ultimate disposal is achieved through direct discharge, landfilling, land application and permanent lagoons.

The focus of the remainder of this text will be to examine the operational and economic feasibility of the coagulant recovery process as a conditioning step

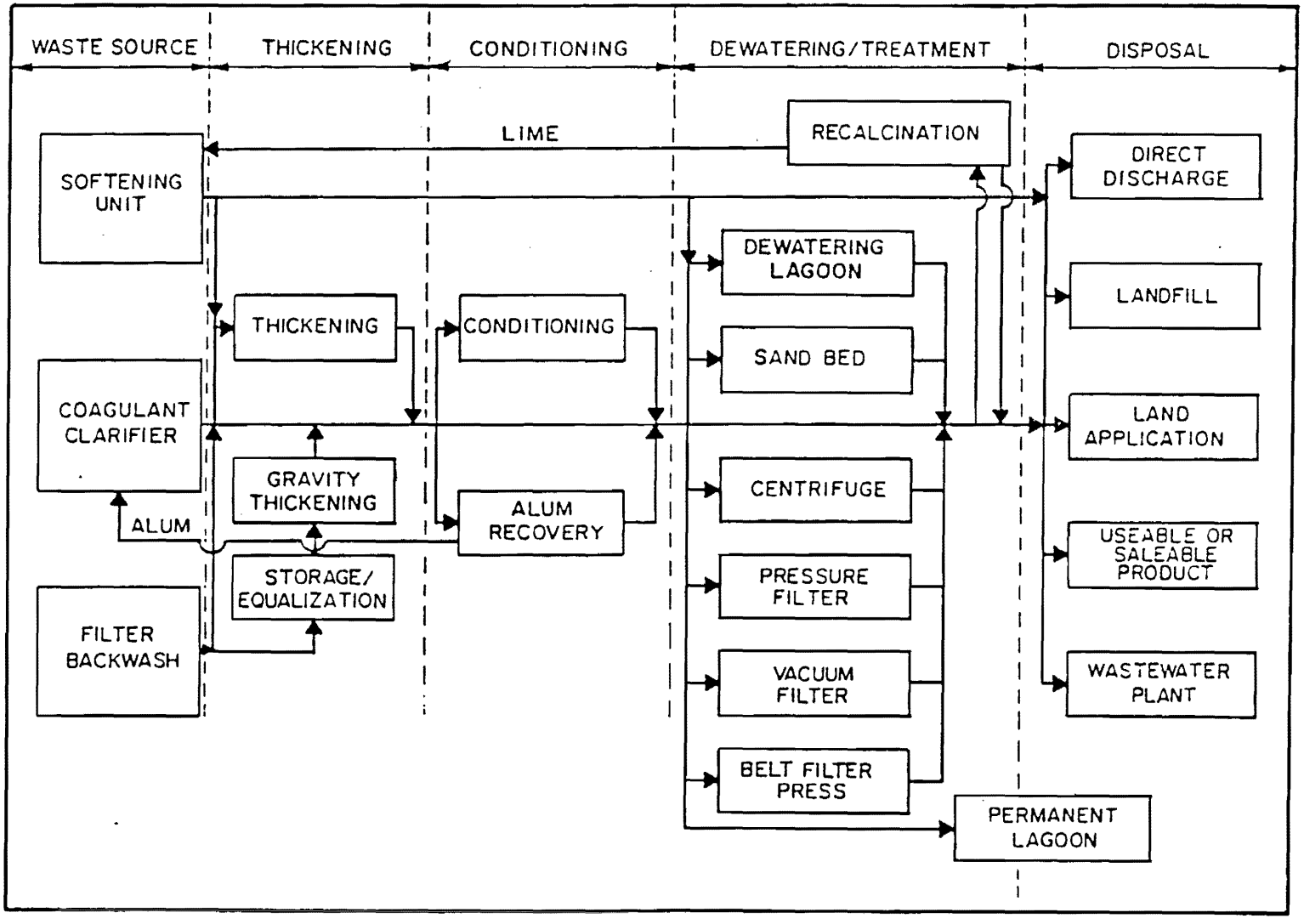

FIGURE 5. Sludge Handling Options for Thickening, Conditioning, Dewatering and Ultimate Disposal of Water Treatment Plant Sludges (Cornwell et al. 1987)

for the minimization of sludge mass and volume for ultimate disposal. Furthermore, the effectiveness of the resulting recovered coagulants will be examined to determine the impact of its use on finished water quality and plant operations.

ALUMINUM RECOVERY FROM WATER TREATMENT PLANT SLUDGES

The recovery of aluminum from alum sludges is a well known process, dating back as early as 1903 in the United States (Roberts and Roddy 1960). The practice later found widespread use in Japan, Great Britain and Poland (Chen et al. 1976). Although acidic extraction using sulfuric acid is the most widely accepted process, use of other acids and bases has been explored. Sulfuric acid, hydrochloric acid, sodium hydroxide and calcium hydroxide have all been considered for use in the recovery of aluminum from alum sludges.

Referring to the pC-pH diagram for aluminum presented in Figure 1, soluble aluminum concentrations increase dramatically with a change in pH from the point of minimum solubility. Therefore, when an aluminum hydroxide sludge in the predominant form of $Al(OH)_3 \cdot 3H_2O$ is conditioned with acid or alkali, aluminum is dissolved to form intermediate polymerization products and hydrated aluminum hydrolysis products. When reused as a coagulant and neutralized to a neutral pH during coagulation, these soluble products will provide benefits similar to those of commercial alum products.

ALUMINUM RECOVERY BY ACIDIC EXTRACTION

Although variations are encountered, the process of acidic aluminum recovery is generally conducted in a rapid mix unit which is followed by a separator, as presented in Figure 6. Thickened sludge is pumped into a rapid mix tank where it is mixed with acid. Acidified sludge is transferred to a separator where supernatant liquid containing aluminum will be recovered to be used as a coagulant in water treatment. Sludge from the underflow of the separator may or may not be neutralized prior to being dewatered. Solids from the dewatering unit are typically disposed of in a sanitary landfill and the resulting liquid is reused as a coagulant, returned to the treatment plant, or discharged to a sewer.

Variables Affecting Acidic Extraction of Aluminum

Two distinct sources of aluminum are present in an alum sludge produced in water treatment: aluminum solids from coagulant addition and aluminum

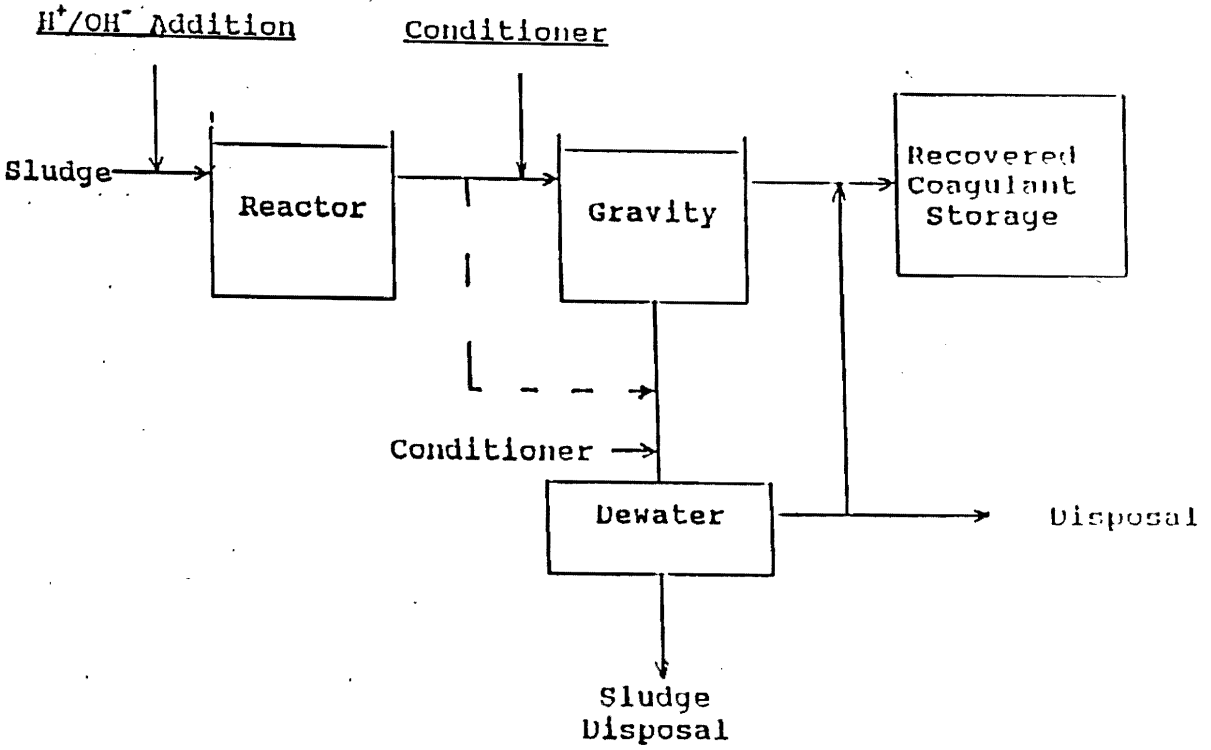

FIGURE 6. Typical Coagulant Recovery Process Diagram for Acidic and Alkaline Extraction of Aluminum and Iron from Water Treatment Plant Sludges

associated with raw water solids, such as clay. Aluminum present in clays is typically in the form of aluminum oxides ($Al_2O_3 \cdot nH_2O$) and is much more resistant to acidification than the aluminum hydroxides produced in coagulation. The dissolution of these aluminum oxides is analogous to the extraction of aluminum from raw material such as bauxite. The process by which aluminum is extracted from bauxite for production of commercial alum utilizes elevated temperatures, high acid doses and extended acid contact for efficient dissolution of the aluminum oxides. Nitric-acid and other digestion procedures in Standard Methods (APHA 1985), which are typically used for determination of total metal contents, are also capable of dissolution of a majority of the Al_2O_3 due to the elevated temperatures and the amount of acid used in these procedures. Therefore, reported total aluminum concentrations in water treatment sludges measured by nitric-acid and other digestions (APHA 1985) include aluminum from coagulant addition [$Al(OH)_3 \cdot 3H_2O$] and a majority of the aluminum from the raw water solids ($Al_2O_3 \cdot nH_2O$).

Sulfuric-acid recovery off aluminum from alum sludges is much less rigorous than nitric acid digestion for analytical determinations of total metal content, with lower acid doses, considerably lower temperatures and a shorter period of contact with sludge solids. Under the conditions for coagulant recovery, aluminum oxides associated with raw water solids may not be dissolved. Investigations of aluminum recovery from aluminum-anodizing sludges, composed primarily of $Al(OH)_3 \cdot 3H_2O$ formed following neutralization of aluminum anodizing wastewaters, indicates that 100 percent dissolution of aluminum hydroxide can be expected using stoichiometric acid additions (Saunders and Shaw 1988). Complete dissolution was obtained at the first measured time increment of 30 min following acid addition, indicating that complete dissolution of $Al(OH)_3 \cdot 3H_2O$ occurred rapidly. The coagulant recovery process operates at room temperature and near stoichiometric acid additions. Under these conditions a majority of the aluminum oxides associated with raw water solids may not be dissolved and will result in less than 100 percent extraction of total aluminum.

Based on the above, 100 percent aluminum dissolution should not be expected in the acid extraction process. Furthermore, 100 percent recovery of aluminum may not be required to achieve recovery of all of the aluminum added as a

coagulant. Therefore, less than 100 percent recovery does not necessarily indicate poor performance of the acid extraction procedure.

Various operational parameters have also been found to have a significant impact on the degree of aluminum dissolution and the feasibility of acidic aluminum recovery. These parameters include: type of acid; stoichiometry of aluminum dissolution; pH of acidified suspensions; and, kinetics of extraction. As will be discussed later, these parameters impact the concentration of aluminum in the recovered coagulant, as well as its quality and utility for reuse as a coagulant in water treatment.

Acid Type: H_2SO_4 vs HCl

Both sulfuric and hydrochloric acids have been investigated for use in coagulant recovery. The reaction between sulfuric acid and aluminum hydroxide may be written as follows:

$$2Al(OH)_3 \cdot 3H_2O + 3H_2SO_4 \Rightarrow Al_2(SO_4)_3 + 9H_2O \quad (1)$$

while the reaction between hydrochloric acid and aluminum hydroxide may be written as:

$$Al(OH)_3 \cdot 3H_2O + 3HCl \Rightarrow AlCl_3 + 6H_2O \quad (2)$$

The stoichiometric sulfuric acid requirement is 1.5 mole/mole Al, 1.1 kg/kg $Al(OH)_3 \cdot 3H_2O$ or 5.45 kg/kg Al, while the stoichiometric hydrochloric acid requirement is 3.0 mole/mole Al, 0.80 kg/kg $Al(OH)_3 \cdot 3H_2O$ or 4.06 kg/kg Al. These stoichiometric acid requirements are based on complete dissolution of solid aluminum species, represented by $Al(OH)_3 \cdot 3H_2O$, to soluble aluminum as Al. The existence of aluminum hydrolysis products can influence this relationship in that the required acid demand would be lower. For example, dissolution of aluminum to the soluble form of $Al(OH)_2^+$ would require only 1/3 of the acid demand discussed previously for dissolution to Al. Therefore, the acid demand can be significantly impacted by the distribution of soluble aluminum species following acid addition.

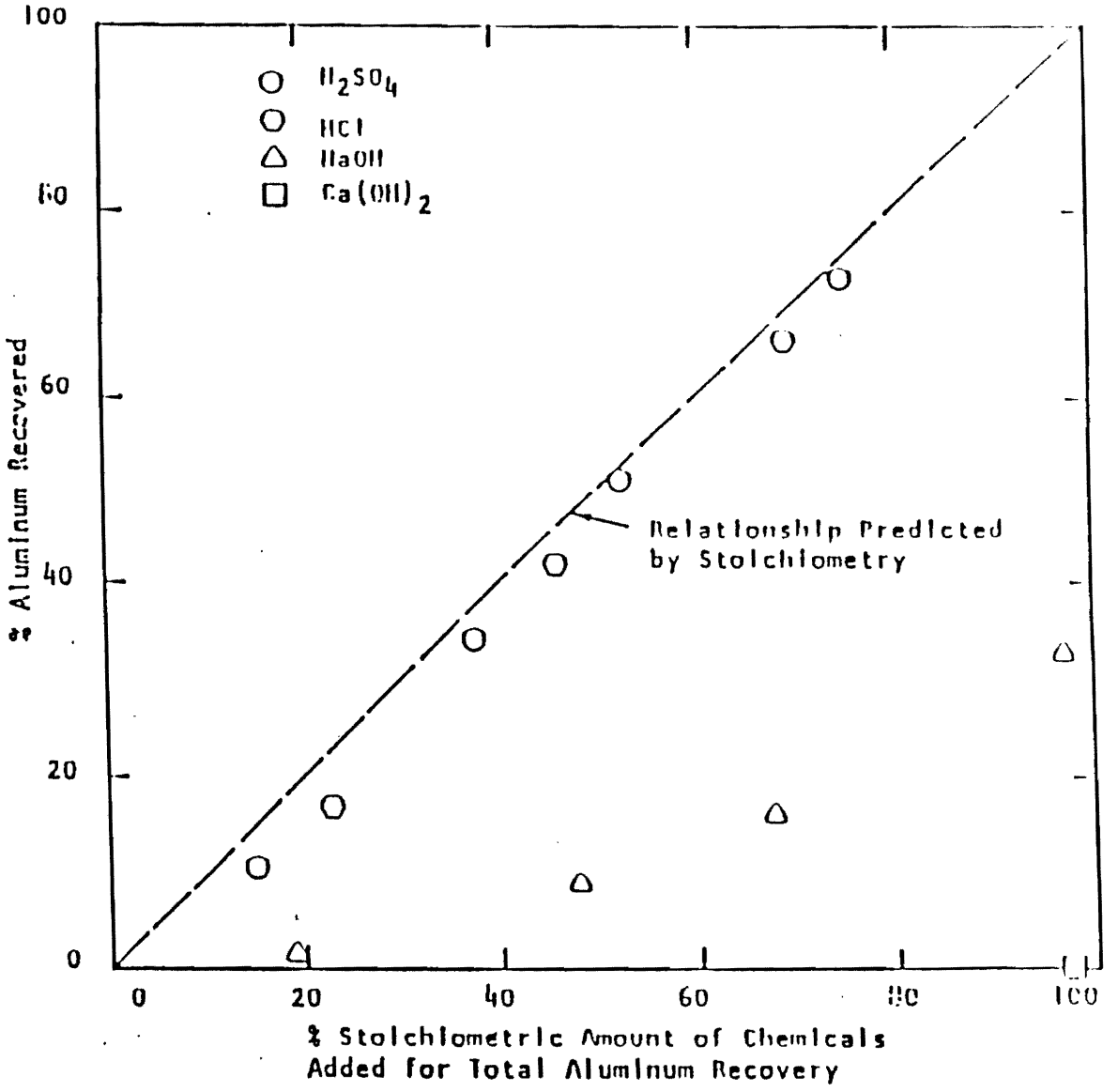

FIGURE 7. Comparison of Experimental and Theoretical Recovery of Aluminum Based on Dissolution of $Al(OH)_3 \cdot 3H_2O$ to Al^{+3} Using Sulfuric Acid, Hydrochloric Acid, Caustic Soda and Hydrated Lime (King et al. 1975)

As presented in Figure 7, King et al. (1975) found stoichiometric predictions were followed closely in examining aluminum recovery as a function of percent of stoichiometric acid addition based on aluminum content. Although HCl was as effective in the recovery process as H_2SO_4, its costs were prohibitively higher, therefore making it less economically advantageous. A majority of the investigations in the literature used sulfuric acid exclusively.

Dissolution of Aluminum in Alum Sludges

Sulfuric acid (H_2SO_4) has been investigated extensively for use in acidic alum recovery from water treatment plant sludges. The stoichiometric acid requirement for dissolution of $Al(OH)_3 \cdot 3H_2O$ to Al^{+3} is 1.5 mole/mole aluminum, 1.11 kg/kg $Al(OH)_3 \cdot 3H_2O$ or 5.45 kg/kg Al. Complete aluminum dissolution is typically not obtained primarily for two reasons.

First, many other acid demanding materials are present in the sludge along with the aluminum solids produced during coagulation. As presented earlier, iron oxides and hydroxides can be present in a sludge sample at concentrations equivalent to aluminum, while manganese and other trace metals are also present in significant quantities. Therefore, if acid addition is based on aluminum content of the sludge, only a fraction of the total aluminum could be dissolved if other acid-demanding materials are present. Secondly, the form of aluminum in alum sludges, i.e., $Al(OH)_3 \cdot 3H_2O$ or $Al_2O_3 \cdot nH_2O$, can have a dramatic impact on aluminum recovery when raw water suspended solids are high. As discussed previously, 100 percent aluminum recovery may not, therefore, be required to recover 100 percent of the aluminum added as a coagulant.

Stoichiometric Evaluation of Experimental Sulfuric Acid Additions

The amount of sulfuric acid added, as well as its concentration, varied widely in experimental procedures due to differing sludge characteristics and a need for accurate measurement in small-scale testing procedures. In most cases, acid doses were normalized to the amount of aluminum recovered from the sludge (i.e., kg H_2SO_4/kg Al recovered) for comparison to stoichiometric acid demands discussed earlier. A more accurate means of comparison would be to normalize acid dose to total aluminum present in the sludge sample. However, in most

instances, the total sludge aluminum content was not known. The acid dose normalized to recovered aluminum will always be higher than the acid dose normalized to total aluminum, thereby artificially inflating the normalized acid dose reported. However, these acid doses normalized to recovered aluminum concentrations will be employed herein for comparison to stoichiometric demands with this in mind.

Laboratory Scale. In comparing aluminum dissolution to the molar ratio of acid and total aluminum present in a sludge sample, Cornwell and Susan (1979) found an optimum acid dose of 1.5 mole/mole Al (5.45 kg/kg Al) for four sludges. As presented in Figure 8, dissolution of 65 to 90 percent of the total aluminum was obtained using this acid dose. With the exception of one sludge, acid doses up to 2.5 mole/mole (9.1 kg/kg Al) did not appreciably increase aluminum dissolution. For example, about 80 percent aluminum dissolution was achieved for the Indianapolis sludge at an acid dose of 5.45 kg/kg Al, with aluminum dissolution only increasing to 85 percent at a dose of 9.1 kg/kg Al.

Isaac and Vahidi (1961) found an optimum acid dose of 3.95 kg/kg aluminum recovered (2.37 kg/kg total Al), where 60 percent of the total aluminum was recovered. During batch testing on a sludge containing 7.75 percent suspended solids with an unknown aluminum concentration, Albrecht (1972) reported using an acid dose of 4.94 kg/m^3 of sludge or 0.64 kg/kg dry solids. Webster (1969) reported using acid doses ranging from 2.41 kg/kg aluminum to 4.14 kg/kg aluminum with recovery of 53 to 81 percent of the total aluminum.

Saunders (1989) reported recovered aluminum concentration as a function of acid addition. As presented in Figure 9, acid additions and recovered aluminum concentrations varied widely among the sludges tested. Maximum recovered aluminum concentrations ranged from 0.566 to 3.436 g/L, while acid doses ranged from about 4 to 11 kg/m^3. Normalization of acid additions to suspended solids concentrations (i.e. kg/kg suspended solids) failed in demonstrating much consistency among the sludges, as shown in Figure 10. Sample D-08-01-S, having the lowest total aluminum concentration (831 mg/L), exhibited the highest normalized acid requirement and was considerably different from the five remaining sludges tested. Examination of the

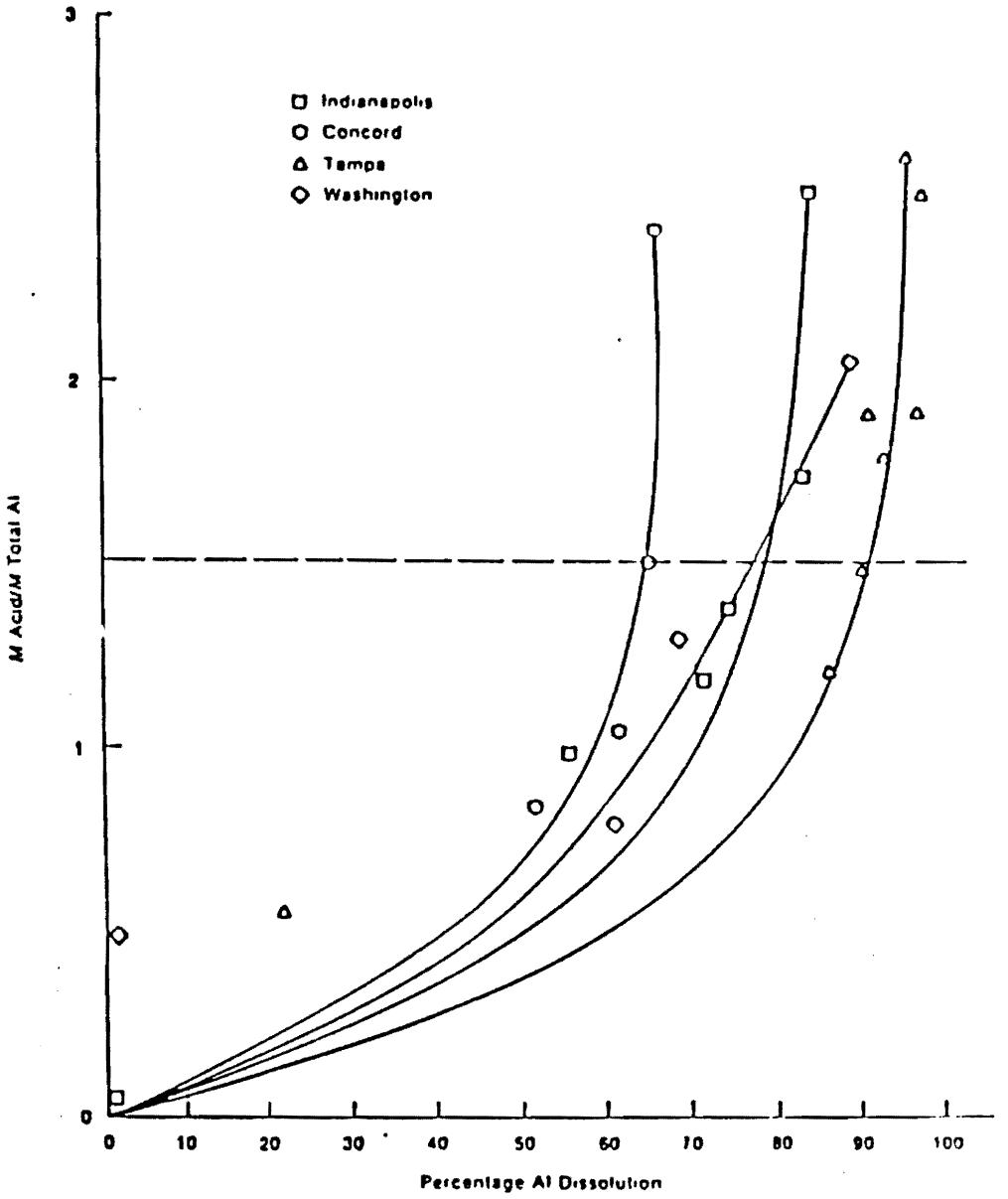

FIGURE 8. Dissolution of Aluminum in Alum Sludges as a Function of Acid Addition (Cornwell and Susan 1979)

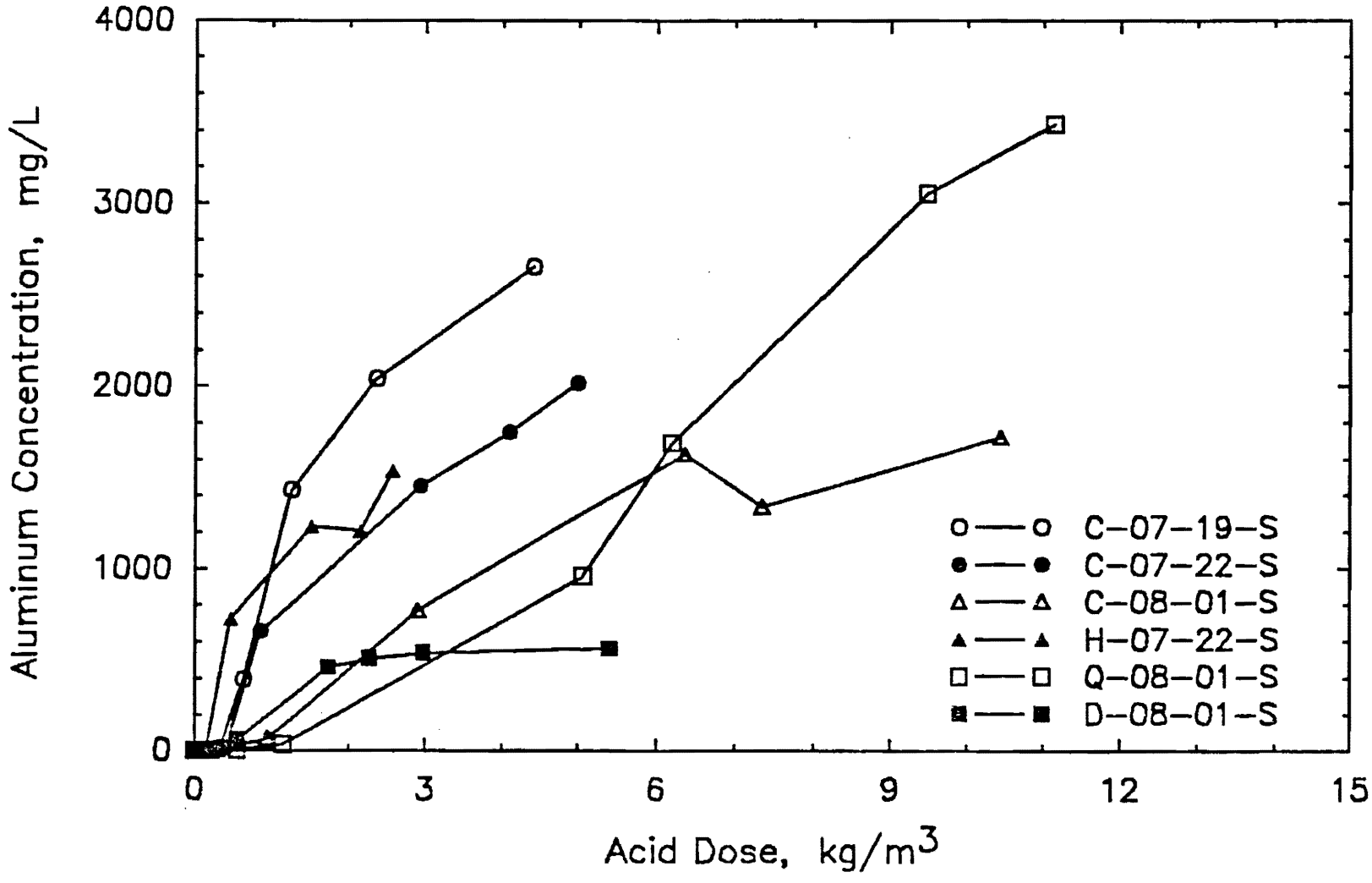

FIGURE 9. Recovered Aluminum Concentration in Supernatant Liquid of Acidified Alum Sludge as a Function of Sulfuric Acid Dose (Saunders 1989)

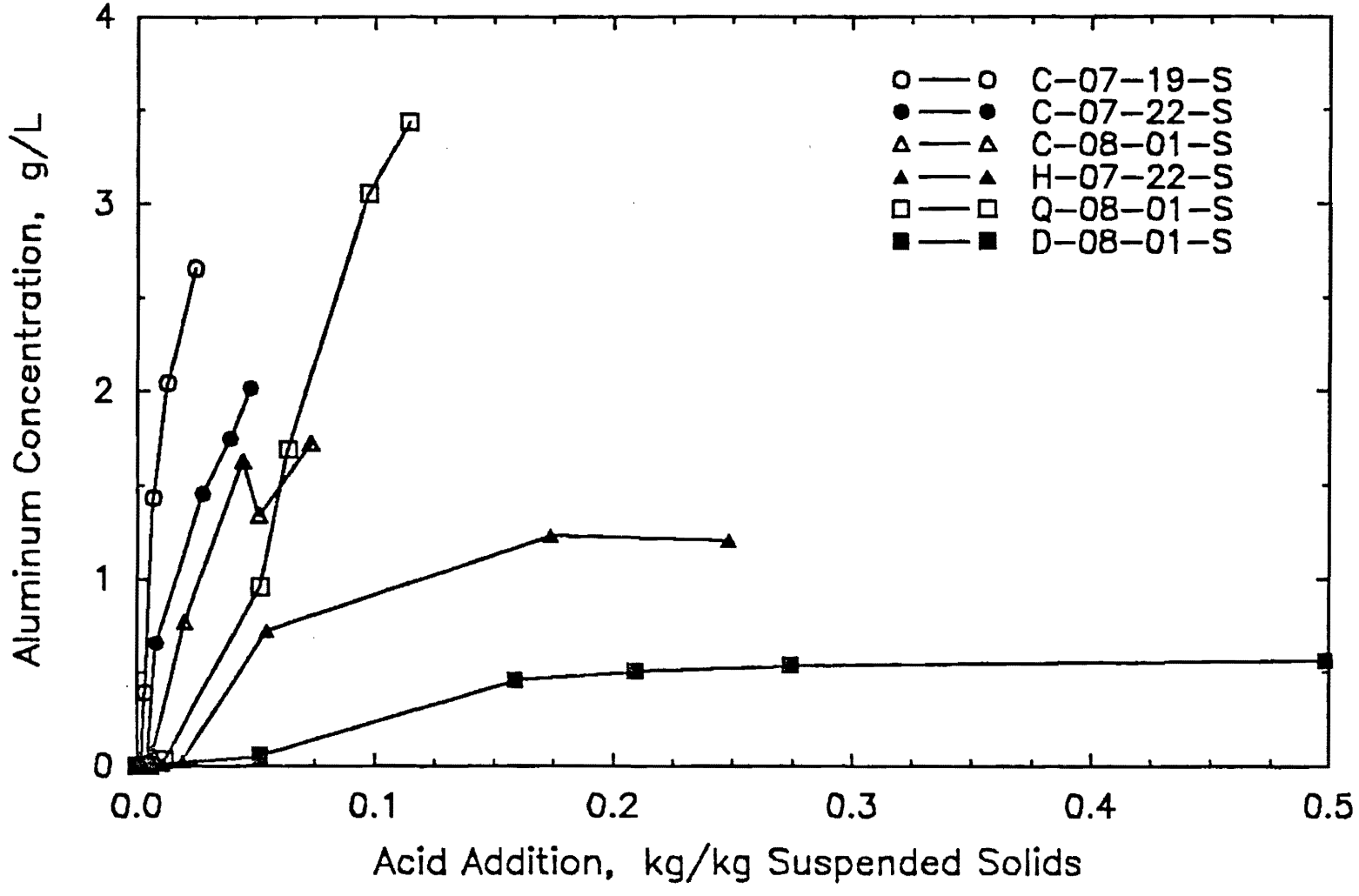

FIGURE 10. Recovered Aluminum Concentration in Supernatant Liquid of Acidified Alum Sludge as a Function of Acid Dose Normalized to Initial Sludge Suspended Solids Concentration (Saunders 1989)

remaining sludges shows little correlation between acid dose and soluble aluminum concentration among the sludges. Although the three Chattahoochee sludges were collected over a two-week period, aluminum recovery varied substantially and normalization with respect to solids concentration failed to show a definite relationship among the data.

Three of the sludges were examined in some detail and will be presented herein. Aluminum recovery was expressed as a function of acid dose normalized to total aluminum content, as presented in Figure 11. The Chattahoochee sludge (C-07-22-S) had a suspended solids concentration of 104.2 g/L and an aluminum concentration of 4.904 g/L, accounting for approximately 23.0 percent of the suspended solids as $Al(OH)_3 \cdot 3H_2O$. A recovered aluminum concentration of 2.016 g/L, 41.1 percent of the total, resulted from an acid dose of 1.0 kg/kg total Al (0.025 kg/kg SS; 4.3 kg/m^3). Based on stoichiometric predictions, this acid dose should result in dissolution of 18.7 percent of the total aluminum, less than half that found experimentally. This would indicate that aluminum is in a soluble hydrolyzed state such as $Al(OH)_2^{1+}$ or $Al(OH)^{2+}$, therefore exhibiting only a fraction of the stoichiometric acid demand described earlier.

The Quarles sludge sample (Q-08-01-S) had a suspended solids concentration of 97.4 g/L and a total aluminum concentration of 5.167 g/L, accounting for approximately 25.9 percent of the suspended solids as $Al(OH)_3 \cdot 3H_2O$. An acid dose of 2.15 kg/kg Al (0.11 kg/kg SS; 11 kg/m^3) resulted in a recovered aluminum concentration of 3.436 g/L, 66.5 percent of the total. Based on stoichiometry as above, 39.6 percent of the aluminum could be recovered as compared to the 66.5 percent experienced experimentally. Again, this appears to indicate aluminum remained in a soluble hydrolyzed form following acidification.

Investigation of the Candler sludge sample revealed that less aluminum was recovered than would be predicted based on stoichiometry alone. The sludge had a suspended solids concentration of 10.85 g/L and a total aluminum concentration of 831 mg/L, which is equal to 37.6 percent of the suspended solids as $Al(OH)_3 \cdot 3H_2O$. An acid dose of 7 kg/kg Al (0.5 kg/kg SS; 5.5 kg/m^3) resulted in a recovered aluminum concentration of 566 mg/L. Stoichio-

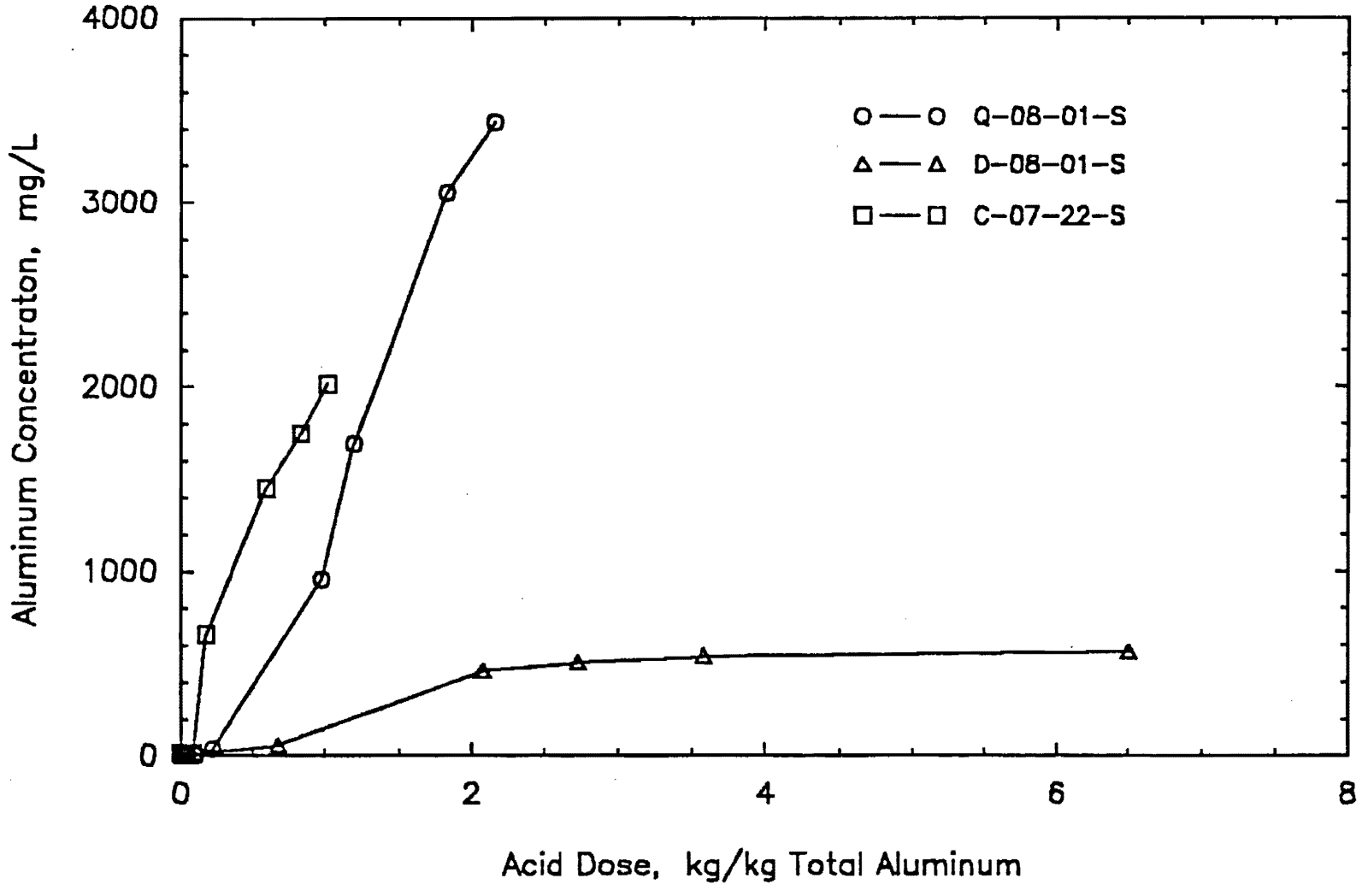

FIGURE 11. Recovered Aluminum Concentration in Supernatant Liquid of Acidified Alum Sludge as a Function of Acid Dose Normalized to Sludge Aluminum Content (Saunders 1989)

metrically, this acid dose is equivalent to 119.5 percent of the total aluminum present, while the actual recovery was only 68.1 percent of the total aluminum. One possible explanation is the presence of other acid demanding materials besides aluminum and the presence of more acid resistant aluminum oxides from the raw water solids.

Pilot Scale. Webster (1969) used the results of laboratory-scale testing to operate a pilot-scale recovery system using an average acid dose of 2 kg/m^3 of sludge over the 10-month period of operation. On one occasion a 5-L sample of sludge was withdrawn from the pilot sedimentation basin for acidification and alum recovery. An acid dose of 1.85 kg/m^3 of sludge was used to obtain a recovered aluminum concentration of 0.45 kg/m^3. This was equivalent to a sulfuric acid dose of about 4.0 kg/kg aluminum recovered.

In pilot-plant testing at Sturgeon Point, Westerhoff and Daly (1974) and Gruninger (1975) reported average acid doses of 0.37 kg/kg sludge solids and 0.43 kg/kg sludge solids for the coagulation-basin and filter-backwash sludges, respectively. Chen et al. (1976) reported aluminum recovery with respect to stoichiometric dose of acid added, as presented in Figure 12. All sludges had an acid demand well aligned with stoichiometric predictions, with aluminum recovery slightly lower than expected in most cases.

Full Scale. Bishop et al. (1987) employed a sulfuric acid dose of 2.0 mole/mole Al (7.27 kg/kg Al) in full-scale testing at Durham, N.C. Large batch acidification using about 0.6 kg/kg dry solids to obtain a pH of approximately 2.0 resulted in 79 and 74 percent recovery of the supernatant having aluminum concentrations of 2.0 and 2.7 kg/m^3, respectively. Since the total aluminum concentration of the raw sludge was not determined, aluminum recovery on a mass basis could not be determined. Roberts and Roddy (1960) reported results of laboratory, pilot and full-scale testing at Tampa. An average acid demand of 4.1 kg/kg aluminum recovered was reported at the optimum operating conditions.

pH of Acidified Suspensions

Acid addition in the extraction process, in many cases, was controlled by

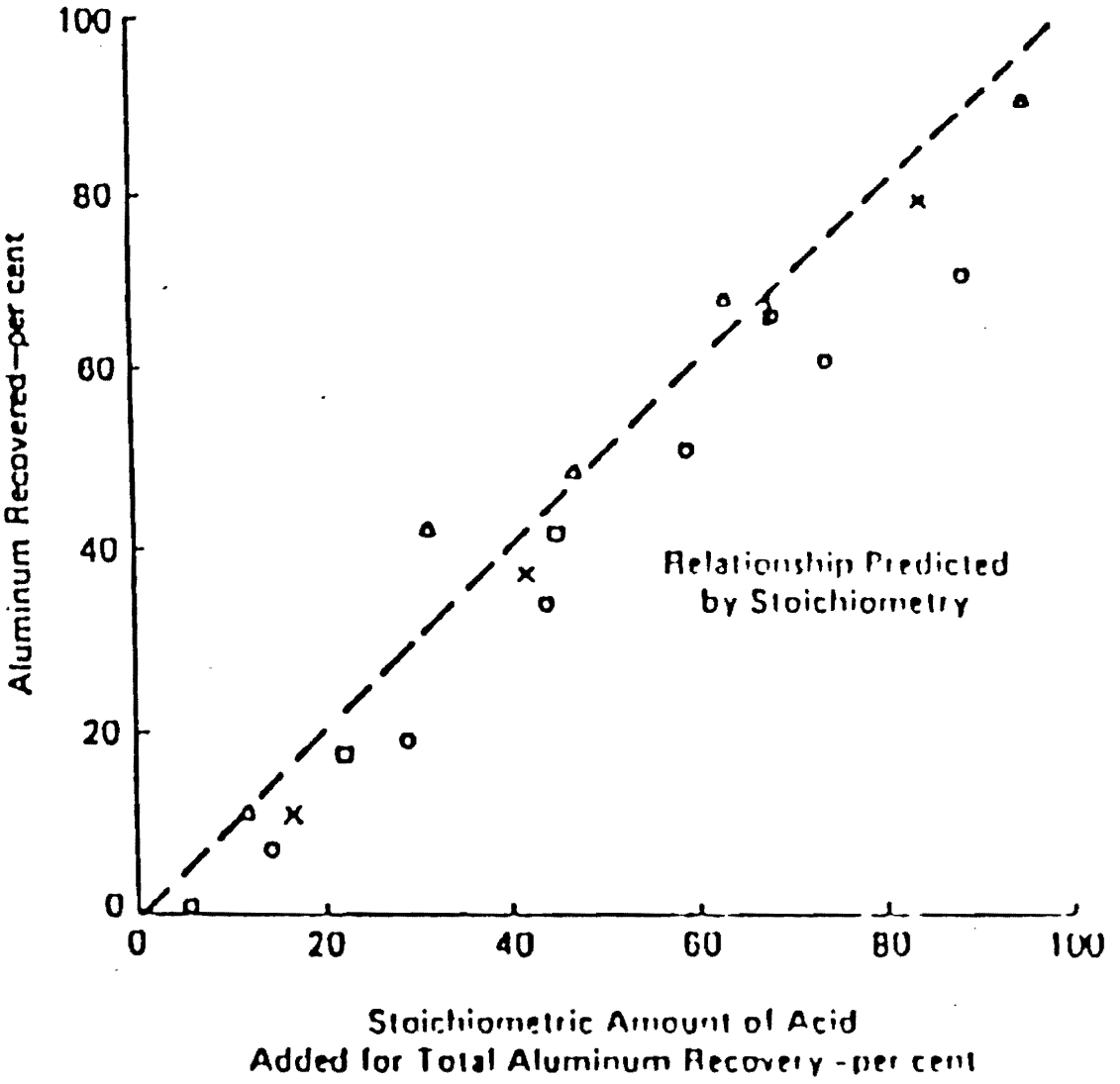

FIGURE 12. Comparison of Experimental Aluminum Recovery Using Sulfuric Acid to the Predicted Stoichiometric Relationship Based on Dissolution of $Al(OH)_3 \cdot 3H_2O$ to Al for Four Alum Sludges Collected in Virginia (Chen et al. 1975)

measurement of suspension pH after acid addition as opposed to addition of stoichiometric acid doses. Suspension pH and aluminum recovery vary widely due to differing sludge characteristics. Chen et al. (1976) concluded that while aluminum recovery could be maximized at a consistent pH for any one sludge, this optimum pH could not be translated to other sludges. The relationship between pH and recovered aluminum for six sludges, as presented in Figure 13 (Chen et al. 1976), is evidence of this fact, demonstrating the variation of aluminum recovery among sludges. However, at a pH value less than about 2.5, recovered aluminum concentrations approached a maximum for a majority of the sludges.

Isaac and Vahidi (1961) concluded that an optimum pH of 3.0 should be maintained. Below a pH of 3.0, color increased dramatically and affected the quality of the recovered coagulant. Roberts and Roddy (1960) concluded that optimum pH varied with raw water alkalinity; a raw water of higher alkalinity requires a lower pH for equivalent aluminum recovery. A summary of pH values and percentage aluminum recoveries reported by numerous investigators are presented in Table 12. Optimum pH was, in most cases, based on maximum recovery potential, product quality and reuse efficiency as a coagulant, as discussed later. The pH values reported in experimental investigations ranged from a value of about 1.0 to 3.0, with recovery of up to 99 percent of the total aluminum.

Saunders (1989) reported pH as a function of acid addition for six sludges collected in Atlanta, GA, as presented in Figure 14. In all cases, an acid dose of less than 1 kg/m^3 lowered the suspension pH to a value of about 4.0. To attain pH values lower than 4, the acid demand varied considerably among the sludges. For example, to reach a pH of 2.0, an acid dose of about 2.45 kg/m^3 was required for the Hemphill sludge (H-07-22-S), while 11.76 kg/m^3 was required to reduce the pH of the Quarles suspension (Q-08-01-S) to a value of 2.0. Attempts at normalization of the data based on suspended solids concentration were not successful, in that much scatter was still evident among the sludges studied. As presented in Figure 15, normalization (i.e., acid addition, kg/kg suspended solids) showed the Candler sludge to have an acid demand of 0.39 kg/kg suspended solids to reach a pH of 2.0, while the remaining sludges were in the range of 0.025 to 0.25 kg/kg suspended solids.

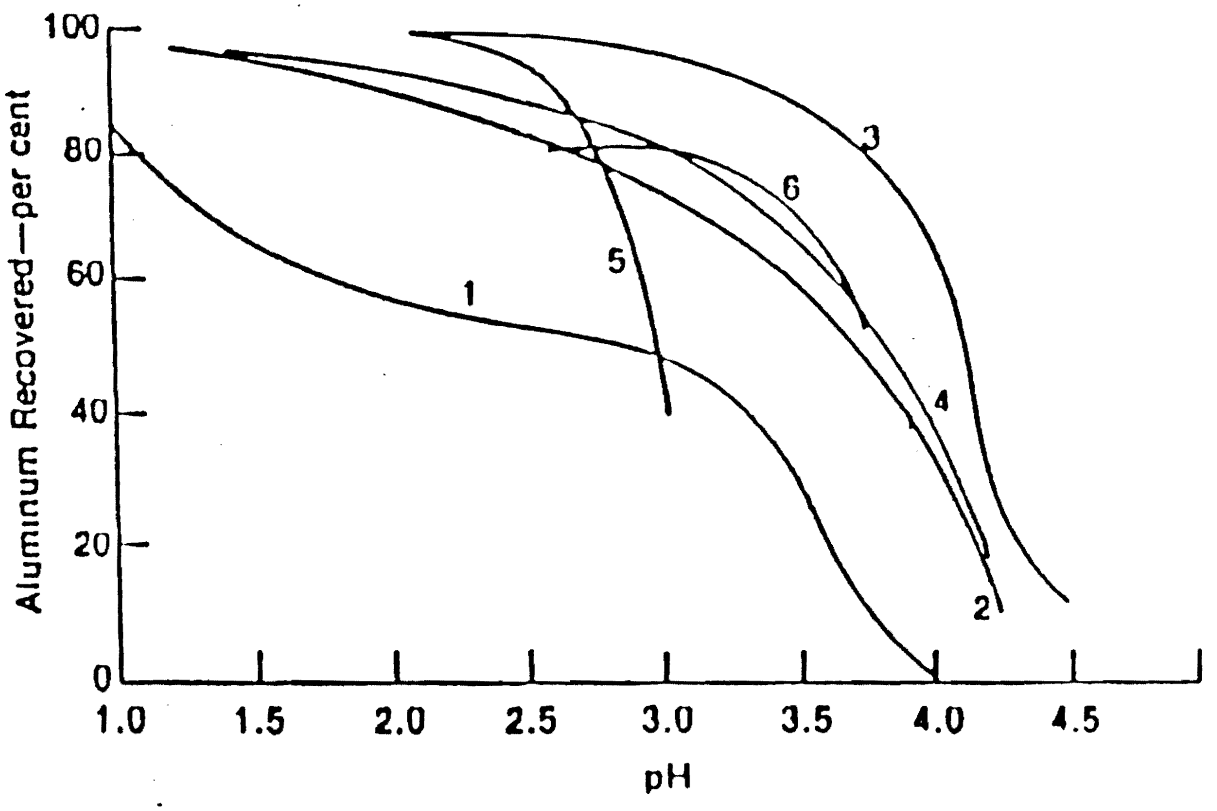

FIGURE 13. Recovery of Aluminum as a Function of Suspension pH of Six Alum Sludges (Chen et al. 1975)

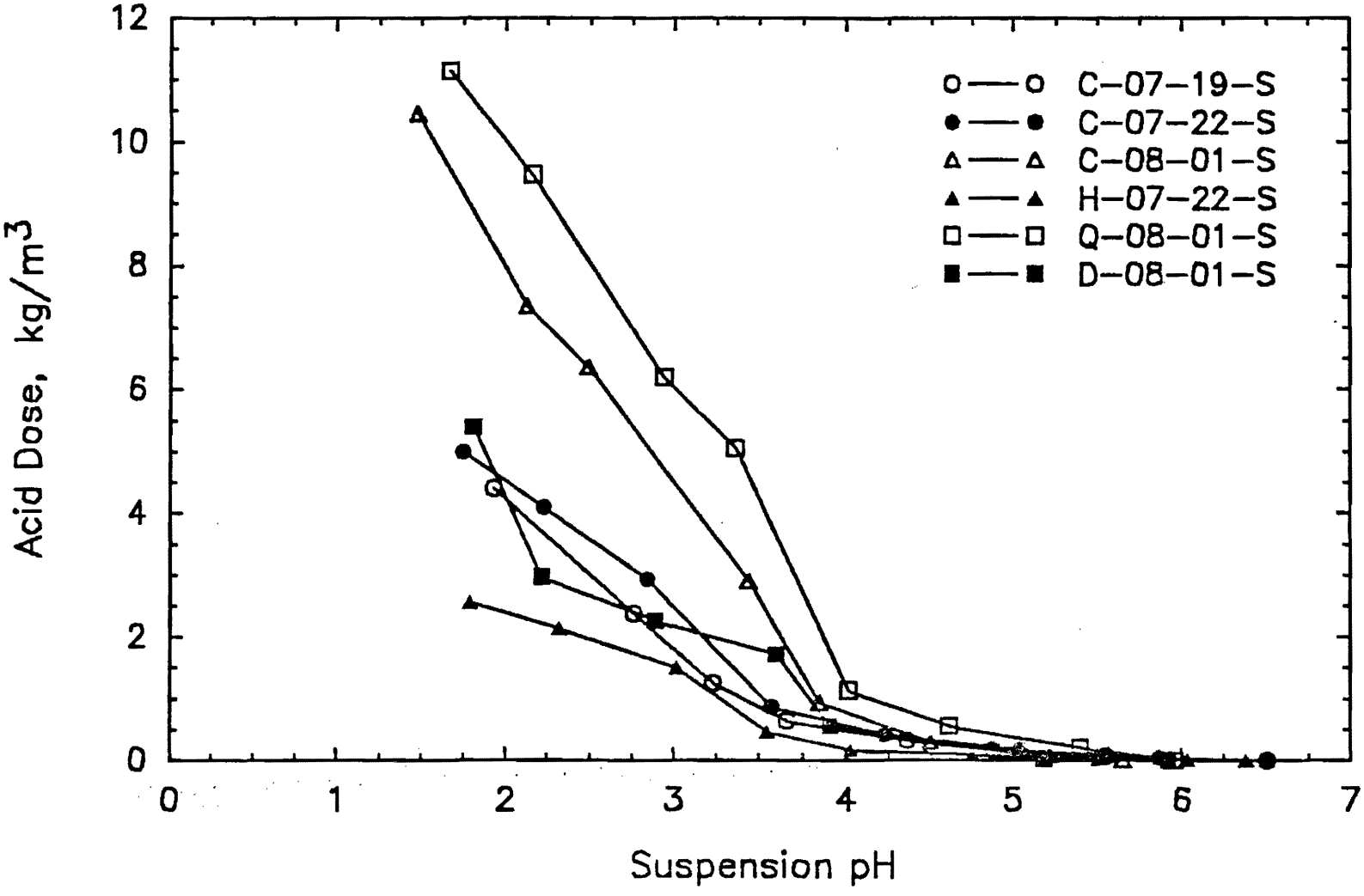

FIGURE 14. Suspension pH for Acidified Alum Sludges as a Function of Sulfuric Acid Dose (Saunders 1989)

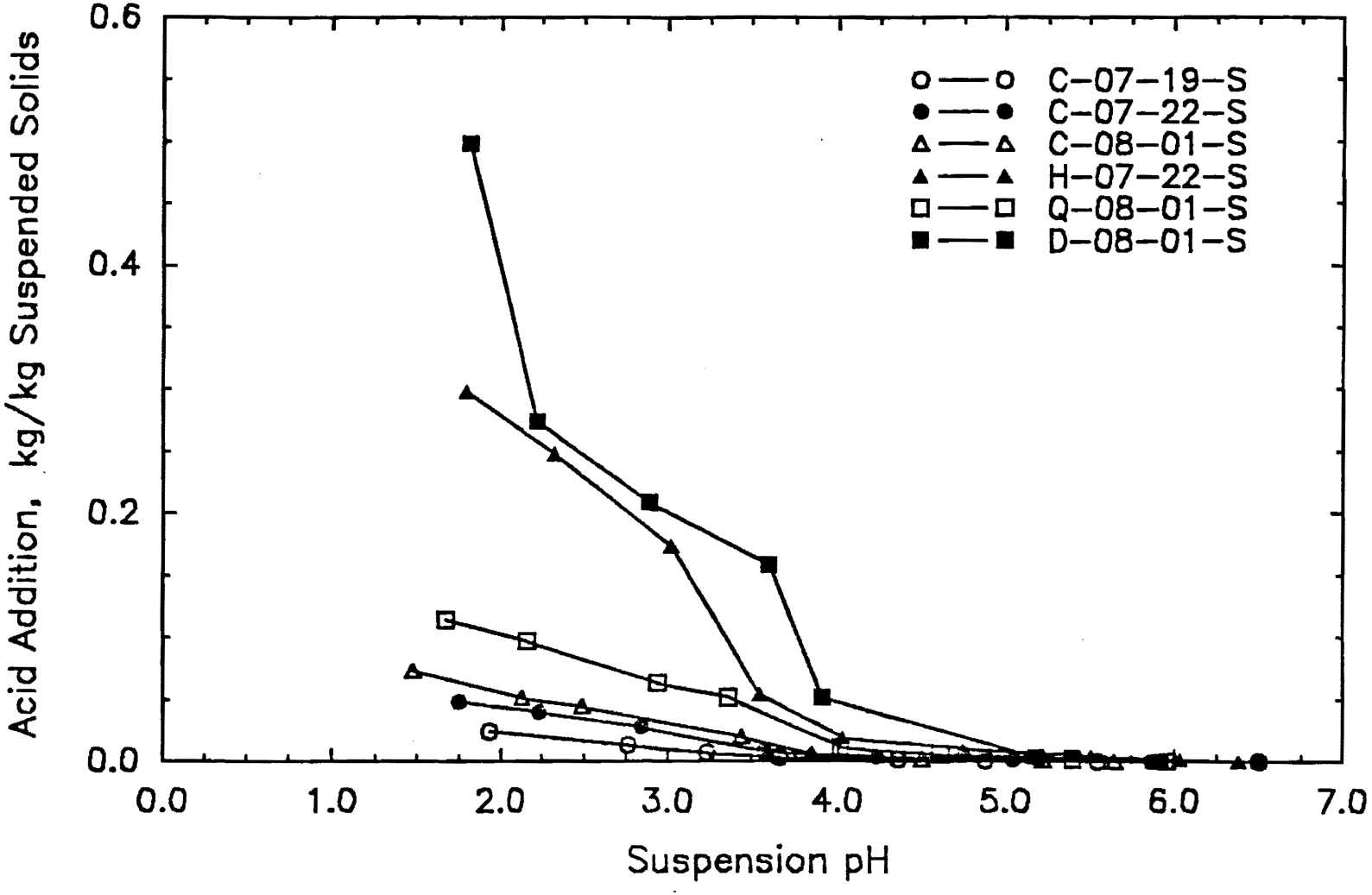

FIGURE 15. Suspension pH of Acidified Alum Sludges as a Function of Acid Dose Normalized to Initial Suspended Solids Concentration (Saunders 1989)

Therefore, it was concluded that the acid dose was not totally a function of suspended solids concentration of the sludge suspensions.

TABLE 12

pH Values Reported for Extraction of Aluminum Using Sulfuric Acid

pH	% Recovery	Location	Reference
1.0	70-99	---	Cornwell and Susan (1979)
1-2	-	Milwaukee	Albrecht (1972)
1.5-2.5	-	Tampa, FL	Roberts and Roddy (1960)
2.0	90	Virginia	King et al. (1975)
2.0	-	Sturgeon Point, NY --- New York State Gainesville, FL	Westerhoff (1973) Fulton (1974) Gruninger (1975) Cornwell and Zoltek (1977)
2.1/2.0	79/74	Durham, NC	Bishop et al. (1987)
2.5	-	Lake Tahoe Scotland	Slechta and Culp (1967) Webster (1969)
3.0	60-65	British Water Works	Isaac and Vahidi (1961)
~2.0		Atlanta, GA	Saunders (1989)

Aluminum concentration as a function of pH for the Atlanta sludges is presented in Figure 16. At a pH of 2.0, recovered aluminum concentrations ranged from about 560 mg/L for the Candler sludge to 3200 mg/L for the Quarles sludge. As presented in Figure 17, normalization of the data to suspended solids concentration shows that the Chattahoochee (C) sludge extracts were fairly well grouped, with aluminum concentrations ranging from 10 to 18 kg/kg suspended solids. Since the Chattahoochee sludge samples were collected over a two-week period, characteristics of the sludges would be similar if the raw water remained of about the same quality, therefore contributing to these similarities.

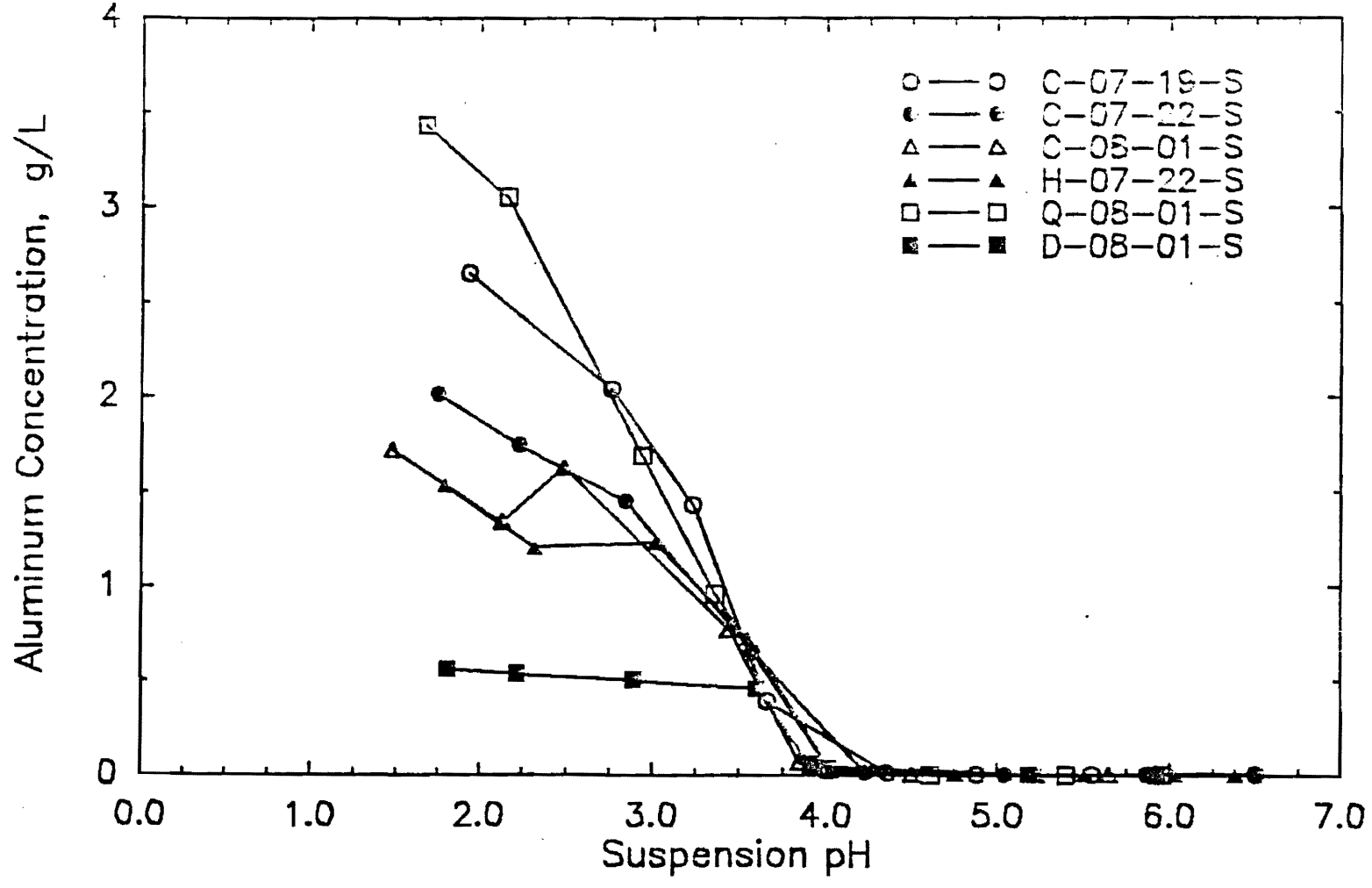

FIGURE 16. Recovered Aluminum Concentration in Supernatant Liquid of Acidified Alum Sludges as a Function of Suspension pH (Saunders 1989)

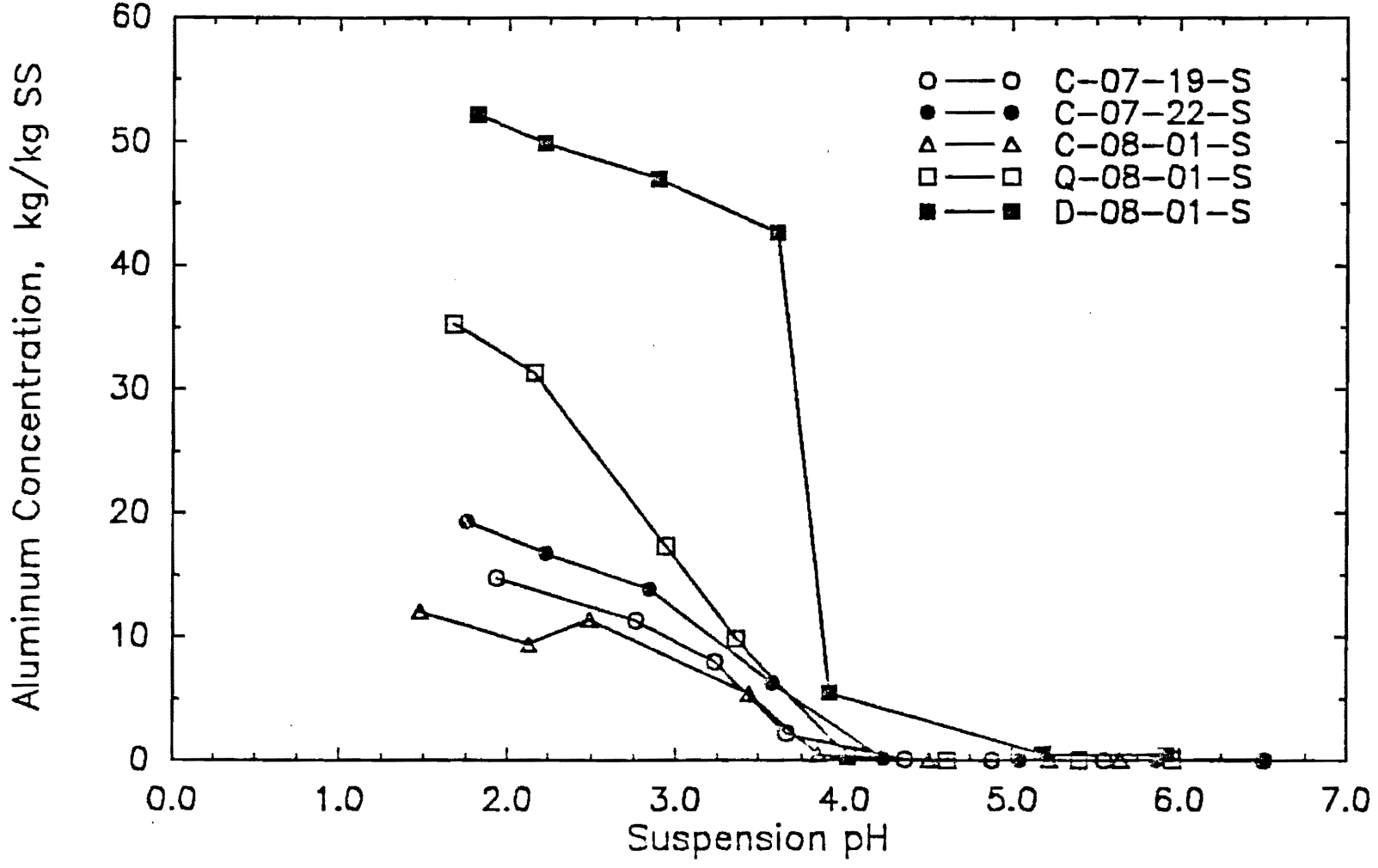

FIGURE 17. Recovered Aluminum Concentration in Supernatant Liquid of Acidified Alum Sludge Normalized to Initial Sludge Suspended Solids as a Function of Suspension pH (Saunders 1989)

Reaction Kinetics of Acidic Extraction

Kinetic tests were performed by Isaac and Vahidi (1961) and Cornwell and Susan (1979) to determine the optimum contact time for the acidic dissolution of aluminum. Both concluded 15 min of contact time were sufficient to obtain equilibrium following stoichiometric acid addition. As presented in Figure 18, Cornwell and Susan (1979) found that approximately 5 min of detention time were required to reach about 80 percent equilibrium, while 100 percent equilibrium was obtained in less than 20 min. Equilibrium was defined by Cornwell and Susan (1979) as the point where no further aluminum was dissolved.

Reported detention times employed in the recovery process ranged from 10 min in full-scale operation in Japan (Westerhoff and Daly 1974) to 30-45 min by Saunders (1989), as presented in Table 13.

TABLE 13

Time of Extraction Reported in Practice for Recovery of Aluminum Using Sulfuric Acid

Time (min)	Scale of Testing	Sludge Source	Reference
10	Full	Japan	Westerhoff and Daly (1974) Goldman and Watson (1975)
25	Pilot	Sturgeon Point	Westerhoff and Daly (1974) Gruninger (1975)
30	Laboratory	England	Isaac and Vahidi (1961)
30	Laboratory	Virginia	King et al. (1975)
30-45	Laboratory	Atlanta	Saunders (1989)

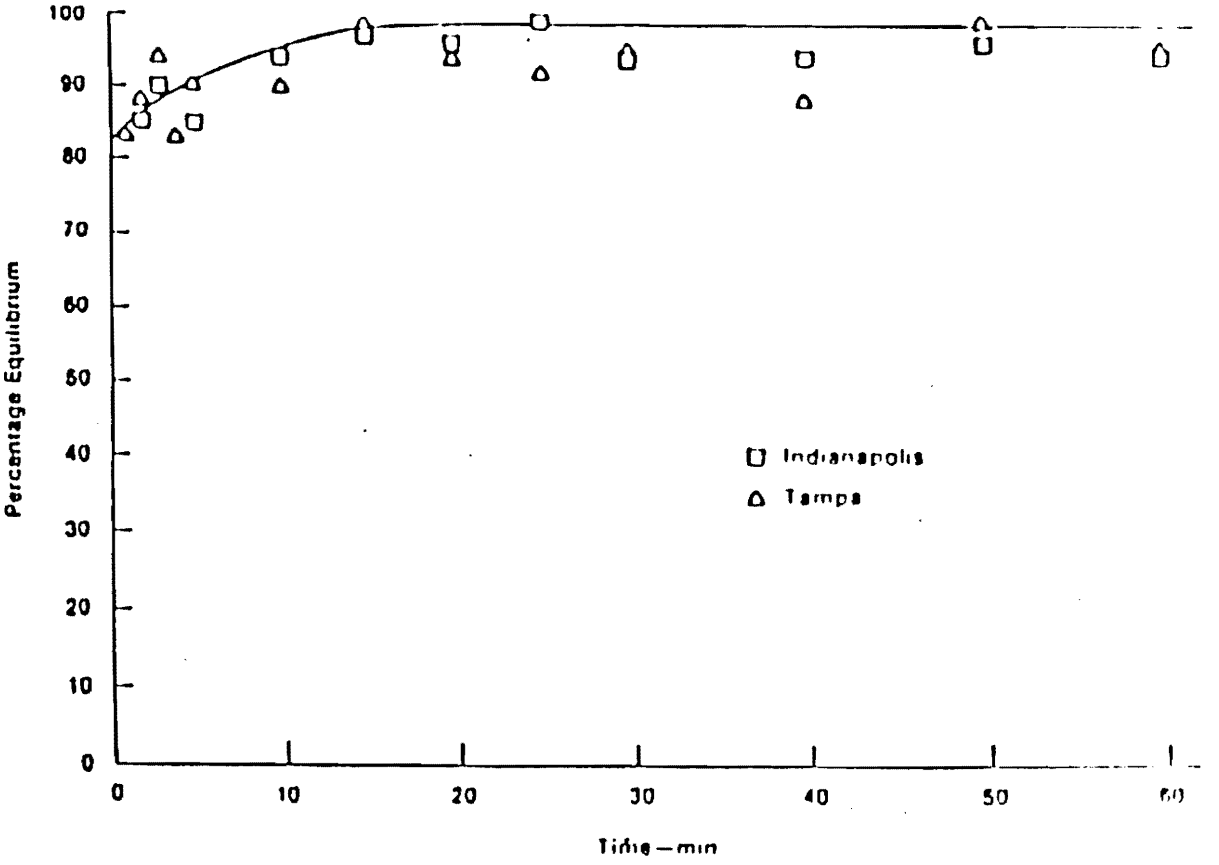

FIGURE 18. Aluminum Dissolution as a Function of Time for Acidified Alum Sludges at an Unspecified pH Value. Equilibrium Aluminum Concentration at an Unknown Time was Used to Establish Percentage EQuilibrium (Cornwell and Susan 1979)

Quality and Utility of Recovered Coagulants

Quality of the products produced by acidic extraction of aluminum from alum sludges was evaluated using aluminum content; organic matter, color, manganese, and iron; and trace organic and inorganic contaminants. Aluminum content is important for use of the product as an effective coagulant in removing color and colloids in the treatment process. Contaminants are of concern since they may be recycled to the treated water during the treatment process as opposed to being removed as part of the sludge solids. Trace contaminants found in recovered products were compared to those found in commercial coagulants in a few instances, providing detailed information on the relative quality of the recovered coagulants.

Utility of acid-extracted coagulants was often tested using a standard jar-test apparatus and by reuse during full-scale testing. Organic matter, color and turbidity of the coagulated waters were used as a measure of coagulation effectiveness in many instances, while some full-scale investigations employed bacteriological, physical and chemical characteristics of the treated waters as a measure of coagulation effectiveness.

Aluminum Content

As described earlier, commercial aluminum sulfate coagulants typically contain about 50 to 60 g/L of aluminum (i.e., approximately 8.25 percent as Al_2O_3). The aluminum concentration in recovered supernatant liquid as reported by various investigators ranged from 360 mg/L to 3700 mg/L (Table 14) and was more than one order of magnitude lower than that reported for commercial coagulants. Westerhoff (1973) reported the recovered coagulant to be more dilute than commercial alum by a factor of 15. Webster (1969), Isaac and Vahidi (1961) and Saunders (1989) reported supernatant aluminum concentration as a function of pH. Webster (1969) reported from 360 to 545 mg/L of aluminum (56 to 85 percent of the total sludge aluminum) in supernatant liquids following acidic recovery of a sludge containing 0.64 percent solids and approximately 644 mg/L of total aluminum (Table 15).

TABLE 14

Aluminum Concentrations for Raw Sludges and Sludge Extracts Reported by Numerous Investigators

Dry Sludge Solids percent	Aluminum Concentration mg/L		Source
	Raw Sludge	Supernatant Liquid	
1.54	2570	740-1590	Isaac and Vahidi (1961)
0.64	644	360-545	Webster (1969)
4-6	--	3700	Gruninger (1975)
1-3	--	2380	Gruninger (1975)
4	--	2330	Gruninger (1975)
2.4	--	2000	Bishop et al. (1987)
3.35	--	2700	Saunders (1989)

TABLE 15

Supernatant Aluminum Concentration as a Function of pH for Extraction of Aluminum from Alum Sludges Using Sulfuric Acid

Supernatant Liquid mg/L as Al	pH	Source
360	3.77	Webster (1969)
434	3.65	Webster (1969)
473	3.50	Webster (1969)
526	3.21	Webster (1969)
523	2.98	Webster (1969)
545	2.64	Webster (1969)
607	3.40	Isaac and Vahidi (1961)
1197	2.95	Isaac and Vahidi (1961)
1444	2.20	Isaac and Vahidi (1961)

Isaac and Vahidi (1961) reported aluminum concentrations of 607 to 1444 mg/L in supernatant liquids recovered from a sludge containing various amounts of aluminum (Table 15). Recovery of aluminum during multiple recycles was also reported by Isaac and Vahidi (1961). During the six cycles reported, aluminum concentrations in the recovered supernatants ranged from about 850 to 900 mg/L.

Saunders (1989) reported recovered aluminum concentrations as a function of acid addition and pH as presented earlier in Figures 9 and 16, respectively. Recovered aluminum concentrations at a pH of 2.0 ranged from 560 mg/L to about 3200 mg/L for the six sludges studied. Soluble aluminum concentrations remained essentially zero until the pH approached a value of 4.0, increasing nearly linearly as the pH decreased. Two of the extract samples exhibited a plateau below a pH value of about 2.5, while the remainder of the extract aluminum concentrations continued to increase. This would indicate the possibility for further aluminum recovery at lower pH values (i.e. higher acid doses).

Fulton (1974) determined that aluminum concentration was also dependent on the suspended solids concentration prior to acidification. Fulton (1974) concluded sludge concentrations from gravity thickeners prior to acidification should be greater than 2.0 percent solids to obtain a sufficient aluminum concentration suitable for coagulation.

Coagulation Effectiveness of Recovered Coagulants

Color and turbidity have been used in laboratory-scale systems to determine the effectiveness of recovered coagulants, while finished water from pilot and full-scale systems was analyzed for bacteriological, physical, and chemical characteristics. Critical coagulation concentration as a function of alum dose was used by Chen et al. (1976) as a measure of alum coagulation effectiveness. Critical coagulation concentration, CCC, was established by extrapolation of the steepest part of a residual turbidity versus alum dose curve to zero turbidity, as illustrated in Figure 19. Critical coagulation concentration as a function of raw water turbidity is presented in Figure 20 (Chen et al. 1976). For both raw waters examined, recovered alum had a

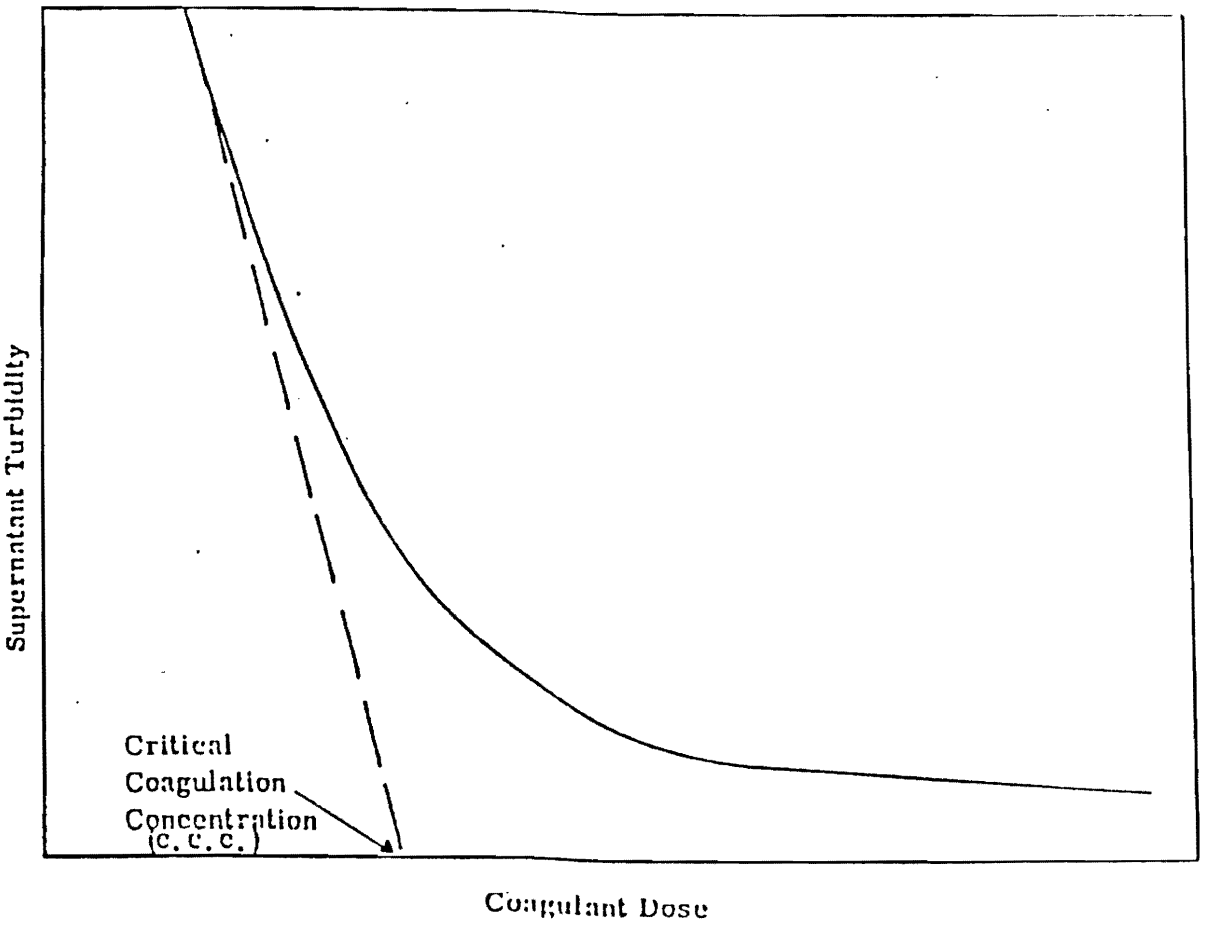

FIGURE 19. Supernatant Turbidity as a Function of Coagulant Dose and Procedure Used to Identify Critical Coagulation Concentration - CCC (Pigeon 1976)

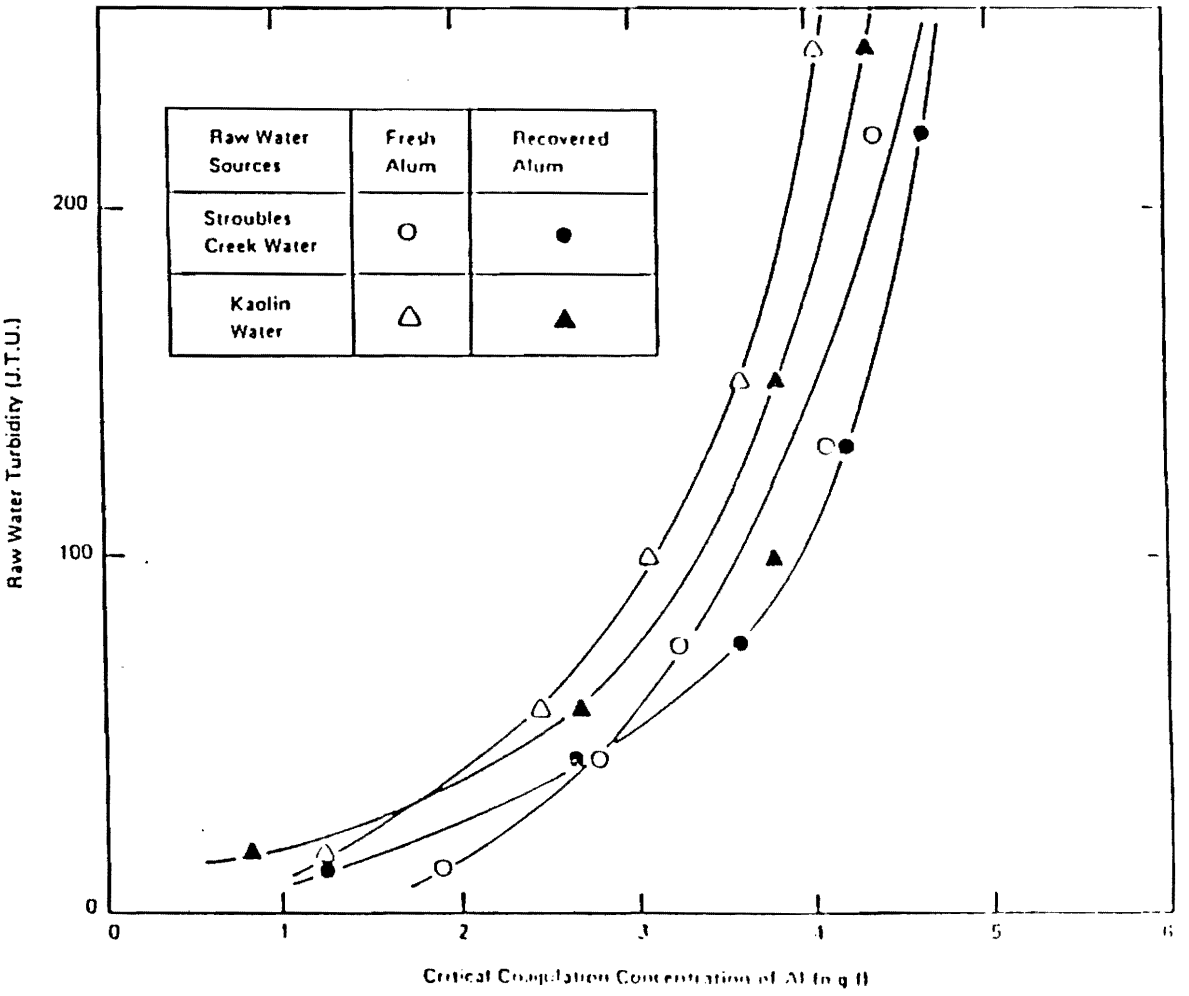

FIGURE 20. Critical Coagulation Concentration as a Function of Raw Water Turbidity for Natural and Artificial Raw Water Suspensions Using Commercial and Recovered Coagulants (Chen et al. 1976)

slightly higher CCC than did commercial alum at high turbidities (>50 JTU), while at lower turbidity, recovered coagulant appeared to be at least as effective as commercial alum.

Slechta and Culp (1967) reported on alum recovery with up to ten recycles of the recovered alum. Although the measure of effectiveness was not reported, the authors concluded that recovered alum was as effective as commercial alum after up to 10 extraction cycles. Isaac and Vahidi (1961) found that 10 mg/L of commercial alum removed 90 percent of raw water color, while equal doses of recovered alum removed only 70 to 80 percent. It was also noted that as the pH of the recovered alum solution dropped below 3.0, the effectiveness of the recovered alum deteriorated.

Saunders (1989) reported on the reuse of recovered coagulants from sludges collected at four water treatment plants in Atlanta, GA. A natural surface water was coagulated with increasing doses of recovered coagulants. As presented in Figure 21, all four coagulants exhibited similar levels of effectiveness, but were not compared to commercial coagulants to determine relative effectiveness. Turbidity was decreased from 11 to 16 NTU initially to less than 1.0 NTU in all cases. The optimum coagulant dose ranged from about 0.8 mg/L to 2.0 mg/L as aluminum, equivalent to alum doses of 8.9 mg/L to 22.2 mg/L. Turbidity increased dramatically at higher aluminum doses indicating some restabilization of particles. This could be due to overdose of coagulant or the depressed pH of the coagulated water attributable to the acidic nature of the recovered coagulants.

Westerhoff (1973) reported extensively on the quality of finished water from pilot- and full-scale plants, both using the same raw water source (Tables 16 and 17). Turbidity of settled and filtered waters from the pilot-plant was higher than that of the full-scale plant. It was not clear if this was caused by the use of recovered alum or due to settling limitations of the pilot-scale plant. It was concluded that there was no significant difference between the two plants and no build-up of impurities was experienced throughout the duration of the study.

In full-scale testing at Durham, N.C., Bishop et al. (1987) reported water

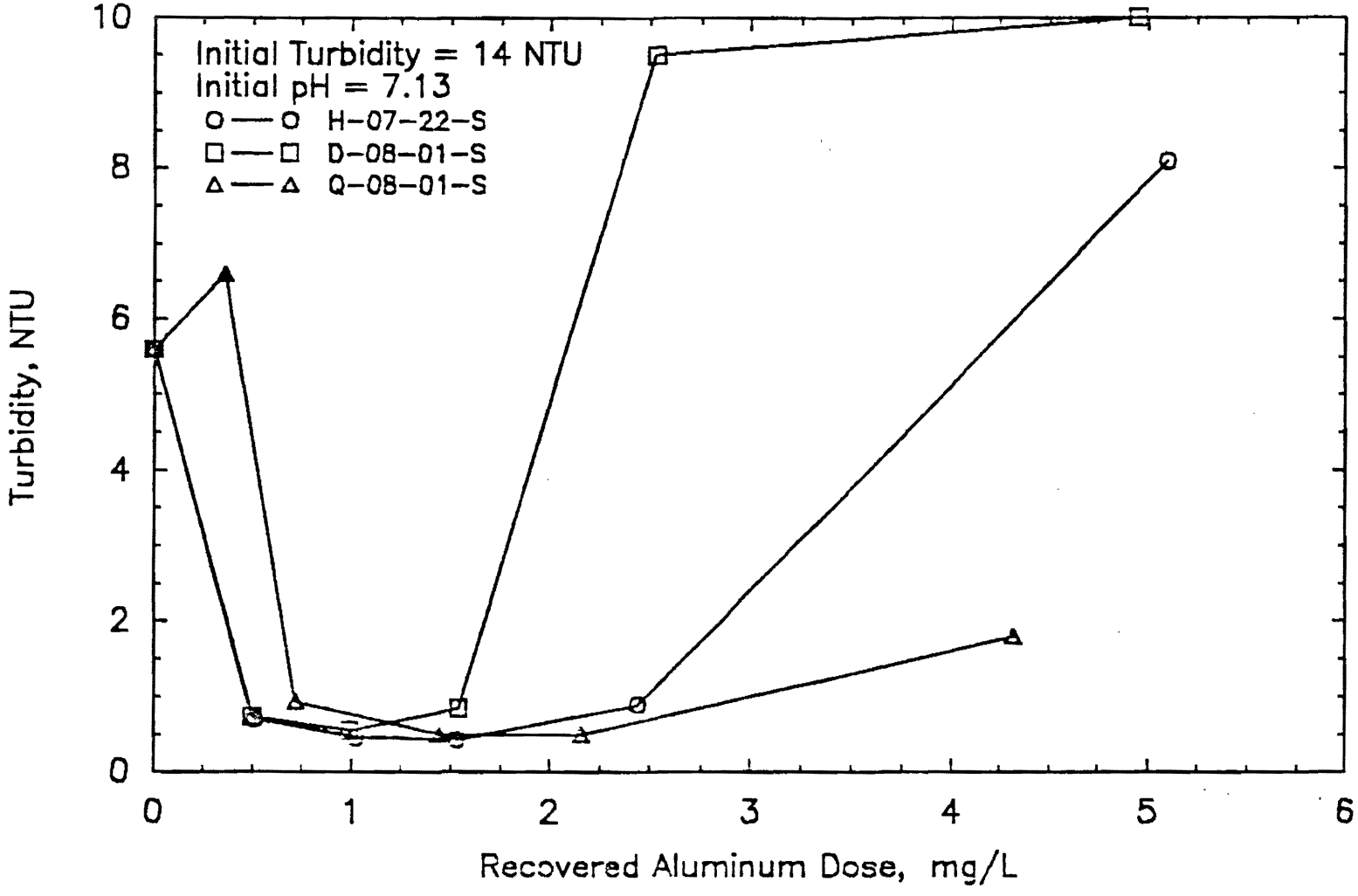

FIGURE 21. Jar Test Results for Chattahoochee River Water Using Recovered Coagulants Produced by Acidification of Alum Sludges Obtained from Chattahoochee-River Water Treatment Plants (Saunders 1989)

TABLE 16

Finished Water Turbidity, Color, Total Iron and Aluminum Concentrations at Sturgeon Point During Pilot-Scale Testing of Coagulant Recovery with a Comparison to Finished Water from the Full-Scale System (Westerhoff 1973)

	Raw Water[1]	Pilot-Plant Filtered Water			Sturgeon Point Filtered Water
		Recovered	Commercial	Total[2]	
pH	8.00	-	-	-	-
Temperature	53.6 F	-	-	-	-
Turbidity, STU	5.9	0.28	0.14	0.22	0.14
Color	1.5	0	0	0	0
Iron, mg/L	0.28	0.04	0.03	0.035	0.02
Aluminum, mg/L	0.01	0.04	0.02	0.03	0.02

[1]The pilot- and full-scale plants used the same raw water source.
[2]Average of all runs conducted using recovered and commercial coagulants in the pilot-plant.

TABLE 17

Finished Water Metal Concentrations During Pilot-Scale Testing of Coagulant Recovery at Sturgeon Point Compared to Full-Scale Finished Water Metal Concentrations (Westerhoff 1973)

Metal	Raw Water[1]	Pilot-Plant[2] Filtered Water	Sturgen Pt. Full-Scale[3] Filtered Water	MCL[4]
Arsenic, mg/L	<0.01	<0.01	<0.01	0.05
Barium, mg/L	<0.1	<0.1	<0.1	1.0
Cadmium, mg/L	<0.001	<0.001	<0.001	0.01
Chromium, mg/L	<0.002	<0.002	<0.002	0.05
Copper, mg/L	<0.1	<0.2	<0.1	1.0
Lead, mg/L	<0.02	<0.02	<0.04	0.05
Manganese, mg/L	<0.1	<0.1	<0.1	0.3
Selenium, mg/L	<0.01	<0.01	<0.01	0.01
Silver, mg/L	<0.02	<0.02	<0.02	0.05
Zinc, mg/L	<0.03	<0.03	<0.02	5

[1]Average of 14 runs
[2]Average of 23 runs
[3]Average of 9 runs
[4]Maximum Contaminant Level, USPHS 1965

treatment plant performance using both commercial and recovered alum coagulants, as presented in Tables 18 and 19. The finished water met all maximum contaminant level (MCL) values and was intrinsically the same for both recovered and commercial coagulants. Settled water turbidity and manganese were both consistently higher on the recovered alum side of the plant, but the filters proved adequate to make the finished waters from each essentially the same. Metal concentrations were monitored closely in test 1, as presented in Table 20. Raw and finished water metal concentrations were compared during periods of commercial and recovered coagulant use. The concentration of metal added to the raw water at the point of coagulant addition attributable to the coagulant are also presented in Table 20. Finished water quality using commercial or recovered coagulant was similar as indicated by finished water metal concentrations. Finished water from the recovered coagulant side of the plant had metal concentrations equal to or lower than the commercial alum side in nearly every instance, meeting all maximum contaminant levels using each coagulant. Iron and manganese, which appeared to be problematic due to high concentrations in the recovered coagulants, were effectively removed in the treatment process. Residual aluminum concentrations in the finished waters were equal in all cases to 400 μg/L. Total organic carbon (TOC) and total trihalomethane formation potential (THMFP) were monitored on finished waters for use as indicators of the impact of organics contained in the recovered coagulant. Average TOC and THMFP values were 5.4 mg/L and 0.214 mg/L, respectively, for the recovered alum and 5.0 mg/L and 0.200 mg/L, respectively, for commercial alum. It was concluded that the overall water quality for the recovered alum side of the plant was equivalent to that found on the commercial alum side.

Results of Repeated Extraction on Recovered Coagulants

As mentioned earlier, Isaac and Vahidi (1961) reported results of repeated recycles of recovered coagulant in pilot-scale testing as presented in Figure 22. Aluminum recovered ranged from 60 to 90 percent of the total, while color of the supernatant liquid fluctuated between 2000 and 4000 Hazen. Although this work was performed using a constant volumetric acid dose (volume acid/volume sludge), recovered supernatant pH remained fairly constant near a value of 2.5. Recovered coagulant was used at a dose equal to the amount of

TABLE 18

Water Treatment Plant Performance Using Commercial Alum (CA) and Recovered Alum (RA) in Test 1 of Full-Scale Operation in Durham, NC (Bishop et al. 1987)

	8/15/85		8/16/85[1]		8/16/85[2]		8/17/85		8/18/85	
Parameters Measured	RA	CA	RA	CA	RA	CA	RA	CA	RA	CA
pH										
Mixing Basin	6.1	6.0	6.2	6.2	6.3	6.2	6.1	6.3	6.3	6.1
Filtered			6.1	6.1	6.2	6.1	6.0	6.1	6.0	6.1
Alkalinity, mg as $CaCO_3$/L										
Raw			24	24			24	24	22	22
Filtered			10	10			10	10	12	12
Turbidity, ntu										
Settled			1.4	1.0			1.8	0.90	2.1	0.92
Filter A			0.10	0.05	0.15	0.05	<0.05	<0.05	0.25	0.10
Filter B			0.15	0.05	0.20	0.05	<0.05	<0.05	0.25	0.10
Filter C			0.15	0.05	0.15	0.05	<0.05	<0.05	0.20	0.10
Filter D			0.10	0.10	0.15	0.10	<0.05	<0.05	0.25	0.10
Filtered Color					<5	<5	<5	<5	<5	<5
Filtered Odor	0	0	0	0	0	0	0	0	0	0
Manganese, mg/L										
Raw			0.84	0.84						
Settled			1.20	0.88						
Filtered			<0.01	<0.01				<0.01	<0.01	<0.01
Filtered Iron, mg/L			<0.05	<0.05				<0.05	<0.05	<0.05
TOC, mg/L			4.4	3.6				3.5	3.8	3.2
Filtered TTHMs, mg/L			0.195	0.196			0.230	0.201	0.227	0.191

[1] a.m.
[2] p.m.
RA Recovered alum
CA Commercial alum

TABLE 19

Water Treatment Plant Performance Using Commercial Alum (CA) and Recovered Alum (RA) in Test 2 of Full-Scale Operation in Durham, NC (Bishop et al. 1987)

	10/2/85[1]		10/2/85[2]		10/3/85[1]		10/3/85[2]		10/4/85	
Parameters Measured	RA	CA	RA	CA	RA	CA	RA	CA	RA	CA
pH										
Filtered	5.3	5.5	5.4	5.9	5.2	5.5	5.2	5.4	5.0	5.6
Mixing Basins	5.5	5.6	5.6	5.9	5.5	5.6	5.5	5.9	5.7	5.3
Alkalinity, mg as $CaCO_3$/L										
Raw	16	16	16	16	17	17	17	17	15	15
Filtered	1	3	3	3	2	4	2	3	2	4
Turbidity, ntu										
Raw	8.5	8.5	9.0	9.0	8.4	8.4	8.4	8.?	8.5	8.5
Settled	4.0	3.0		2.0	3.5	2.0	4.5	1.6	4.0	2.5
Filter A	0.45	0.45	0.45	0.10	0.50	0.20			0.30	0.25
Filter B	0.45	0.45	0.50	0.15	0.65	0.25			0.35	0.30
Filter C	0.45	0.40	0.55	0.20	0.60	0.20			0.35	0.25
Filter D	0.40	0.40		0.15	0.40	0.25			0.30	0.30
Filtered Color	<5	<5			<5	<5			<5	<5
Filtered Odor	0	0	0	0	0	0	0	0	0	0
Manganese, mg/L										
Raw	0.2	0.2			0.26				0.04	
Settled	0.70	0.40			0.82	0.54			0.76	0.42
Filtered	<0.01	<0.01			0.03	0.02			0.04	<0.01
Filtered Iron, mg/L	0.09	0.05			0.14	0.04				
				<0.05	<0.05	<0.05				

[1] a.m.
[2] p.m.
RA Recovered alum
CA Commercial alum

TABLE 20

Metal Contaminant Concentrations (μg/L) in Full-Scale Testing at Durham, NC Using Commercial Alum (CA) and Recovered Alum (RA) Coagulants (Bishop et al. 1987)

Metal	Raw Water 8/16	Metals Attributable[1] to Coagulants		Finished Water Metals Concentration					
				8/16/85		8/17/85		8/18/85	
		Commercial	Recovered	RA	CA	RA	CA	RA	CA
Cadmium	<5.0	-	BD	<5.0	<5.0	<5.0	<5.0	<5.0	<5.0
Chromium	<5.0	0.35	0.70	<5.0	<5.0	<5.0	<5.0	<5.0	<5.0
Copper	<5.0	<0.01	0.70	<5.0	<5.0	<5.0	5.8	<5.0	5.7
Iron	310	43.1	341.6	<50	<50	<50	<50	<50	<50
Sodium	5450	2.1	7.72	5570	5800	5780	5800	5760	5800
Potassium	1830	0.21	7.02	1850	1840	1980	1880	1970	1910
Manganese	840	0.06	297.18	10	40	40	40	40	<10
Nickel	<5.0	<0.01	0.07	<5.0	<5.0	<5.0	<5.0	<5.0	<5.0
Lead	<5.0	0.06	0.05	<5.0	<5.0	<5.0	<5.0	<5.0	<5.0
Zinc	<5.0	0.04	2.11	<5.0	<5.0	<5.0	<5.0	<5.0	<5.0
Calcium	5550	0.23	3.28	5600	5400	5600	5500	5550	5400
Magnesium	2650	0.46	6.32	2650	2700	2700	2600	2600	2650
Aluminum	1400	2340	2340	400	400	400	400	400	400
Silicon	5000	0.53	9.83	4600	4800	4950	4800	4500	4200
Barium	85	0.02	0.23	30	35	30	30	35	45
Silver	<5.0	0.01	BD	<5.0	<5.0	<5.0	<5.0	<5.0	<5.0
Arsenic	<5.0	0.11	0.12	<5.0	<5.0	<5.0	<5.0	<5.0	<5.0
Selenium	<5.0	-	BD	<5.0	<5.0	<5.0	<5.0	<5.0	<5.0
Mercury	<0.5	<0.01	0.00	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5

[1]Based on an aluminum dose of 2.34 mg/L as Al
BD Below detection in the coagulant
< Less than reported value
RA Recovered aluminum
CA Commercial aluminum

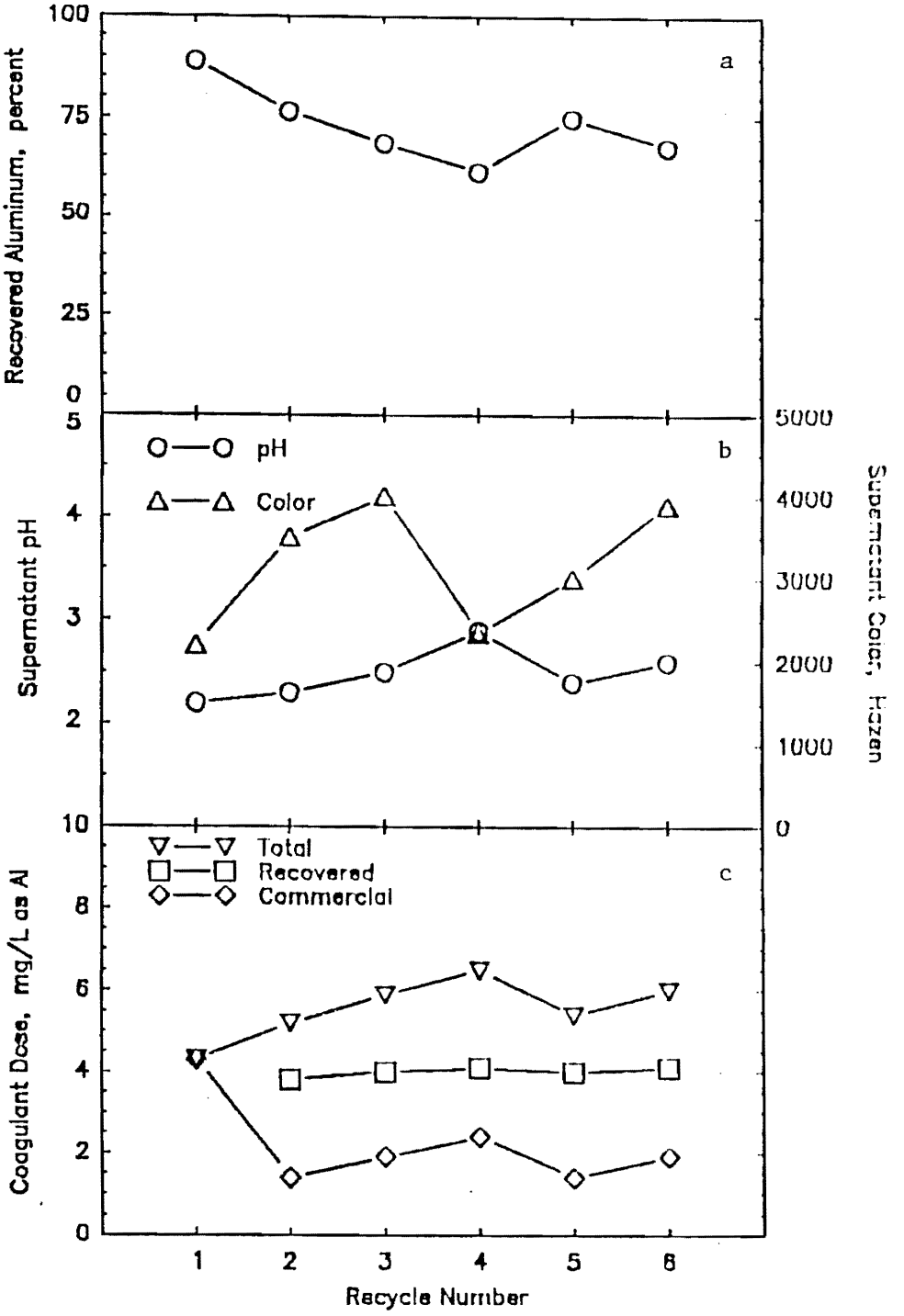

FIGURE 22. Results of Repeated Aluminum Extraction During Pilot-Scale Testing of Aluminum Recovery (Isaac and Vahidi 1961)

a. Percent aluminum recovery using equivalent acid doses for each cycle
b. Resulting supernatant pH and color following acid addition
c. Commercial and recovered aluminum
Dose Required to Maintain Consistent Finished Water Color for Each Batch of Recovered Coagulant

coagulant recovered in a given day (mg/d) divided by the flow rate in the pilot-plant (L/d) to equal the maximum allowable dose (mg/L). Commercial alum was added as make-up to provide consistent color removal from the raw water as determined by jar tests and was typically added at a dose equivalent to one half the recovered coagulant dose.

Although no data were presented to support their conclusions, Roberts and Roddy (1960) reported no change in the chemical or bacteriological quality of the finished water with 8 to 10 recycles of recovered coagulant during experimental testing at Tampa.

Recovered Coagulant Trace Contaminants

Organic Matter and Color. Cornwell and Susan (1979) examined dissolvable and nondissolvable solids fractions of an alum sludge at pH of about 1, as presented in Table 21 for five sludges. The highly varying nature of alum sludges is evident, with no consistency of the fractions observed between sludges. Dissolvable solids ranged from 9 to 86 percent of the total, while the organic portion of the dissolvable solids was from 2 to 26 percent. Aluminum concentrations in the sludges were comparable, but aluminum as a fraction of initial sludge solids concentration varied significantly from 2 to 22 percent. Although no data on color were presented, the authors concluded that an increase in soluble organic solids would cause an increase in the color of the resulting recovered alum.

Isaac and Vahidi (1961) reported the effects of decreasing pH on color of the resulting supernatant liquid as presented in Table 22. The raw sludge used had a total solids concentration of 1.55 percent, total volatile solids of 0.65 percent and a total aluminum concentration of 0.26 percent as Al. It was concluded that for this particular sludge a pH of 3.0 should be maintained, since below this value the color of the recovered supernatant liquid increased dramatically (Table 22).

Supernatant liquid color was also examined over multiple recycles in pilot scale testing by Isaac and Vahidi (1961). As presented in Figure 23, the color increased in the first two cycles to 4000 Hazen, after which it

TABLE 21

Alum Sludge Dissolvable Solids at a pH of 1.0
(Cornwell and Susan 1979)

Parameter	Indianapolis	Concord	Tampa	Moline	Washington
Initial Solids %	1.7	1.7	1.6	1.7	12.1
Total Aluminum Content					
Concentration, mg/L	2400	2400	3500	295	3750
Fraction of Initial Solids	0.14	0.14	0.22	0.02	0.03
Fraction of Initial Solids Dissolved at pH = 1.0					
Dissolvable	0.38	0.54	0.86	0.09	0.35
Inorganic	0.26	0.36	0.61	0.07	0.09
Organic	0.12	0.18	0.25	0.02	0.26

TABLE 22

Recovered Supernatant Color as a Function of Suspension pH
for an Alum Sludge* Conditioned with Sulfuric Acid
(Isaac and Vahidi 1961)

pH	Aluminum Recovery Percent	Supernatant Color Hazen
3.9	7	50
3.15	38	875
3.05	53	1550
3.0	66	1750
2.5	79	5600
2.2	--	7000

*Total sludge solids = 1.55%
Total volatile solids = 0.65%
Total aluminum = 0.26%

fluctuated between 2000 and 4000 Hazen. Fulton (1974) stated that a highly colored product would result from the acidification of a raw sludge containing larger amounts of suspended organic matter. Although no data were presented, Fulton (1974) stated that this color would not be transferred to the finished water, but instead would be removed by the coagulation process with other organic material.

Manganese and Iron. Both manganese and iron are soluble at the low pH values employed in acidic extraction of aluminum. For this reason, they are of concern in the recovery process since they would be present in the recovered alum and could cause a deterioration of the finished water quality. Limitations of 0.05 mg/L and 0.3 mg/L for manganese and iron, respectively, were established as secondary standards (EPA 1975) to prevent black and red water caused by precipitation of manganese oxides and iron hydroxide. Data presented in Table 23 show the variance in concentrations found in commercial and recovered alum by various investigators.

TABLE 23

Iron and Manganese Concentrations Reported in Commercial and Recovered Aluminum Coagulants

Product	pH	Fe, mg/L	Mn, mg/L	Reference
commercial	--	1160	1.7	Bishop et al. (1987)
commercial	--	850	1.8-7.0	Cornwell and Westerhoff
commercial	--	2080	--	Saunders (1989)
commercial	--	1845	--	Saunders (1989)
recovered	2.1-0.25	300-800	--	Wang and Yang (1961)
recovered	2.0	292	255	Bishop et al. (1987)
recovered	3.8-2.6	2.4-14.0	--	Webster (1969)
recovered	2.0	92-1885	18.5-200	Saunders (1989)

Iron and manganese concentrations in commercial products were reported by three investigators. Cornwell and Westerhoff (1982) reported an iron concentration of 850 mg/L, while manganese concentrations ranged from 1.8 mg/L to 7.0 mg/L. Bishop et al. (1987) reported iron and manganese concentrations of 1160 mg/L and 1.7 mg/L, respectively in a commercial coagulant in use at Durham, NC. Saunders (1989) reported iron concentrations of 2080 mg/L and 1845 mg/L in commercial coagulants from Atlanta, while manganese concentrations were not reported.

In general, iron concentrations in recovered coagulants were equal to or less than those found in commercial products. Wang and Yang (1961) reported iron concentrations ranging from 300 to 800 mg/L, while Webster (1969) found 2.4 to 14 mg/L of iron in acid recovered coagulants. Bishop et al. (1987) found an iron concentration of 292 mg/L in coagulant recovered in full-scale testing at Durham. Saunders (1989) reported iron concentrations ranging from 92 to 1885 mg/L in laboratory-scale testing in Atlanta. Manganese concentrations in recovered coagulants were reported by Bishop et al. (1987) and Saunders (1989) to be 255 mg/L and 18.5 to 200 mg/L, respectively. These reported concentrations in recovered coagulants ranged from 1 to 2 orders of magnitude higher than the levels reported in commercial coagulant products.

Isaac and Vahidi (1961) reused recovered coagulants for six cycles to investigate the possible build-up of iron in recovered alum. A constant acid dose (volume acid/volume sludge) was employed, therefore causing a change in aluminum recovery in each cycle. Make-up doses of fresh alum were also added to facilitate consistent color removal for each cycle. As presented in Table 24, iron present in the recovered supernatant liquid decreased through the first four cycles, while remaining constant in the last three, indicating iron concentration did not build-up in the recovered coagulant. The fate of this iron, either as part of the residual sludge solids after extraction or in the treated water, was not examined. Wang and Yang (1961) concluded that iron would be in the ferric state causing the formation of iron flocs during flocculation, which would enhance coagulation.

TABLE 24

Iron Concentration Contained in Recovered Aluminum Coagulants during Repeated Recycle (Isaac and Vahidi 1961)

Cycle	Coagulant Added, mg/L of Al			Recovered Iron mg/L	pH
	Commercial	Recovered	Total		
1	4.3	--	4.3	35.0	2.2
2	1.4	3.8	5.2	28.0	2.3
3	1.9	4.0	5.9	18.2	2.5
4	2.4	4.1	6.5	14.0	2.9
5	1.4	4.0	5.4	14.0	2.4
6	1.9	4.1	6.0	17.5	2.6

Inorganic Trace Contaminants. Bishop et al. (1987) and Saunders (1989) conducted extensive investigations into inorganic contaminants contained in acid extracted coagulants. In both cases, commercial coagulants in use at the facilities were also examined on the same basis for comparison to the extracted coagulants.

Bishop et al. (1987) reported the concentration of inorganic contaminants from acidic extraction of aluminum from alum sludge in full-scale testing at Durham, N.C. Alum sludge was conditioned with sulfuric acid to a pH of 2.1 by addition of 2082 L (3810 kg) of concentrated sulfuric acid to 2994 kg of dry sludge solids. This was equivalent to an acid dose of 2.0 mole/mole of recovered Al (7.27 kg/kg Al), compared to a theoretical demand of 1.5 mole/mole (5.45 kg/kg Al) discussed previously. Metal composition of the recovered coagulant is presented in Table 25 and has been normalized to aluminum concentration (i.e., metal mg/kg Al) for comparison to Water Chemicals Codex (National Research Council 1982) criteria.

TABLE 25

Metals Concentration in Recovered Coagulant During Full-scale Testing at Durham, NC (Bishop et al. 1987)

Metal	mg/L	mg/kg Al	RMIC[1] mg/kg Al
Al	1,970	--	--
Ag	BD	--	330
As	1.1	50	330
Ba	0.3	100	--
Ca	2.8	1,400	--
Cd	BD	--	77
Cr	0.6	300	330
Cu	0.6	300	--
Fe	292	146,000	--
Hg	0.002	0	11
K	6.1	3,000	--
Mg	5.5	2,700	--
Mn	255	127,000	--
Na	6.5	3,300	--
Ni	0.06	30	--
Pb	0.03	20	330
Se	BD	0	77
Si	8.5	4,200	--
Sn	--	--	--
Zn	1.7	900	--

[1]RMIC value calculated using 150 mg/L as alum (13.5 mg/L as Al) and a safety factor of 10 (National Research Council 1982)

The aluminum concentration of the extract was 1970 mg/L and aluminum was the predominant metal found. Iron and manganese were the next largest constituents with concentrations of 292 mg/L and 255 mg/L, respectively. Other metals tested ranged from 0.03 mg/L to 6.5 mg/L. The metal concentrations were normalized to aluminum content for comparison to RMIC values established by Water Chemicals Codex (National Research Council 1982). Arsenic was found at a concentration of 1.1 mg/L, which is equivalent to about 50 mg/kg Al, compared to the RMIC of 30 mg/kg Al. Although the chromium concentration of 300 mg/kg Al (0.6 mg/L) was within the Water Chemicals Codex (National Research Council 1982) specifications (330 mg/kg), these data and others presented later indicate high concentrations of chromium may be expected in recovered coagulants using the acid extraction process, as well as commercial coagulants. Although no RMIC value has been established for iron or manganese, concentrations were notably high at 146,000 mg/kg Al and 127,000 mg/kg Al, respectively.

Saunders (1989) also reported metal concentrations in acidic extracts in laboratory-scale testing of Atlanta sludges. A 4-L volume of sludge solids was contacted with acid for 30 to 45 minutes and the suspensions centrifuged (2700 x g) prior to examination of the clarified centrate for metal composition. Characteristics of the sludge suspensions and acid addition data are presented in Table 26. In general, the suspensions were conditioned to attain a final pH of about 2.0 using acid doses ranging from 0.19 kg/kg SS to 3.4 kg/kg SS. Metal composition (mg/L) and normalized metal composition (mg/kg Al) are presented in Table 27 for the Chattahoochee (C) sludges and Table 28 for the Hemphill (H), Candler (D) and Quarles (Q) sludges. Aluminum concentrations ranged from about 550 mg/L for the Candler extract to 1900 mg/L for the Chattahoochee (C-07-19) sample. Chromium exceeded the RMIC value of 330 mg/kg Al in all extracts, ranging from 640 mg/kg Al to 2140 mg/kg Al. Lead was also observed in high concentrations in the recovered coagulants, ranging from about 600 mg/kg Al to 2160 mg/kg Al, well above the RMIC of 330 mg/kg Al. Additional metals for which RMIC values have been established (i.e., silver, cadmium and mercury) were detected at concentrations well below the RMIC values established by Water Chemicals Codex (National Research Council 1982) specifications.

TABLE 26

Characteristics of Clarified Sludge Extracts Developed for Trace Metal and Organic Analyses by Saunders (1989)

Sample	Date of Extraction	Acid Addition		pH	Solids			
		meq/L	kg / kg SS		Initial Sludge SS kg/m^3	Acidified Sludge TS %	Acidified Sludge TDS %	Acidified Sludge SS %
C-07-19-S	8/21	338	0.19	2.36	179.4	18.59	2.76	15.83
C-07-22-S	8/23	227	0.22	2.51	104.2	-	-	-
C-08-01a-S	8/20	198	0.14	2.15	142.9	14.52	1.62	12.90
H-07-22-S	8/23	288	3.3	1.86	8.67	-	-	-
Q-08-01-S	8/23	342	3.4	1.95	97.4	11.32	2.5	8.82
D-08-01-S	8/23	157	1.4	1.72	10.85	1.88	0.79	1.09

All samples were clarified by centrifugation (2700 x g) and analysed.

TABLE 27

Metals Composition of Clarified Sludge Extracts from Chattahoochee Sludge Samples (Saunders 1989)

	C-07-19		C-07-22		C-08-01a		C-08-01a Filtered	
Metal	mg/L	mg/kg Al	mg/L	mg/kg Al	mg/L	mg/kg Al	mg/L	mg/kg Al
Al	1900	--	1250	--	1250	--	1200	--
Ag	0.0018	0.95	0.0011	0.88	0.0011	0.88	0.0033	2.75
Ba	ND	--	ND	--	ND	--	ND	--
Cd	0.002	1.05	0.01	8.00	0.0169	13.52	0.0165	13.75
Cr	2.78	1463.16	1.71	1368.00	2.67	2136.00	1.07	891.67
Cu	1.67	878.95	2.7	2160.00	2.28	1824.00	1.33	1108.33
Fe	1885	99200.00	1208	966400.00	490	392000.00	458	381666.67
Hg	0.0003	0.16	0.0014	1.12	0.0004	0.32	0.0033	2.75
Mn	200	10500.00	145	116000.00	95	76000.00	80	66666.67
Ni	ND	--	ND	--	ND	--	ND	--
Pb	1.32	694.75	2.7	2160.00	1.24	992.00	1.15	958.33
Sn	ND	--	ND	--	ND	--	ND	--
Zn	5.3	2789.47	4.7	3760.00	4.5	3600.00	4.5	3750.00

ND - Not detected in acid extracts

TABLE 28

Metal Composition of Clarified Sludge Extracts from Hemphill (H), Candler (D) and Quarles (Q) Sludge Samples (Saunders 1989)

	H-07-22		D-08-01		Q-08-01	
Metal	mg/L	mg/kg Al	mg/L	mg/kg Al	mg/L	mg/kg Al
Al	1020	--	550	--	1800	--
Ag	0.005	4.90	0.0011	2.00	0.002	1.11
Ba	ND	--	ND	--	ND	--
Cd	0.007	6.86	0.0163	29.64	0.0169	9.39
Cr	0.852	835.29	0.372	676.36	1.152	640.00
Cu	0.796	780.39	0.525	954.55	3.07	1705.56
Fe	274	268627.45	92	167272.73	1151	639444.44
Hg	0.0036	3.53	0.0014	2.55	0.0005	0.28
Mn	55	53921.57	18.5	33636.36	79	43888.89
Ni	ND	--	ND	--	ND	--
Pb	0.778	762.75	0.479	870.91	1.074	596.67
Sn	ND	--	ND	--	ND	--
Zn	1.6	1568.63	0.5	909.09	2.9	1611.11

ND - Not detected in acid extract

Other metal concentrations were also notably high, although no RMIC values have been established. Iron and manganese were present in concentrations ranging from 99,000 to 990,000 mg/kg Al and 10,000 to 116,000 mg/kg Al, respectively. Copper was present in concentrations ranging from 700 to 2100 mg/kg Al, while zinc concentrations ranged from 900 to 3800 mg/kg Al.

The percentage of the total sludge metal extracted was also determined by Saunders (1989) on four sludge samples collected in Atlanta, GA. As presented in Table 29, the percentage extracted ranged from a low of 3 percent to a high of 100 percent, varying widely with the sludge source. Recovered aluminum varied from 26 percent to 100 percent of the total sludge aluminum, while iron recovery ranged from 18 to 56 percent. Based on these results, only a fraction of the sludge metal content would be present in the recovered coagulants. This would indicate that there is a fraction of the metals which will be wasted from the treatment process with the residual sludge solids, thereby minimizing the build-up of sludge metals in the recovered coagulant products.

Organic Trace Contaminants. The organic composition of the Atlanta sludge extracts was examined by Saunders (1989) using soluble organic carbon (SOC) analysis and humic substance extraction with XAD-8 resin. SOC and humic acid concentrations in the acidified sludge extracts (pH≈2) are presented in Table 30. SOC concentrations ranged from 326 to about 1800 mg/L and were of the same order of magnitude as the recovered coagulant aluminum concentra- tions. The concentration of humic substances ranged from 160 to 1140 mg/L, constituting 38 to 78 percent of the SOC. Soluble organic carbon concentration as a function of pH is presented in Figure 23. As the pH decreased below a value of about 4.0, SOC concentration increased dramatically. Comparison to aluminum dissolution as a function of pH, as presented earlier in Figure 17, indicated that as aluminum began to dissolve with a decrease in pH below a value of 4.0, SOC concentrations increased concomitantly. Organics present in the sludge as adsorbed species on alum flocs or complexed aluminum precipitates were released into solution as aluminum dissolution proceeded, therefore increasing the SOC concentration with a decrease in pH.

TABLE 29

Percentage of Total Sludge Metal Extracted by Sulfuric Acid Addition
(Saunders 1989)

Metal	C-07-22	Q-08-01	D-08-01	H-07-22
Al	26	35	66	(115)
Ag	5	14	40	(125)
Ba	ND	ND	ND	ND
Cd	91	26	(102)	35
Cr	31	19	74	90
Cu	65	62	(127)	(147)
Fe	29	24	18	56
Hg	35	3	50	(213)
Mn	58	63	47	138
Ni	ND	ND	ND	ND
Pb	44	16	86	55
Sn	ND	ND	ND	ND
Zn	36	31	50	100

ND = Not detected in acid extract

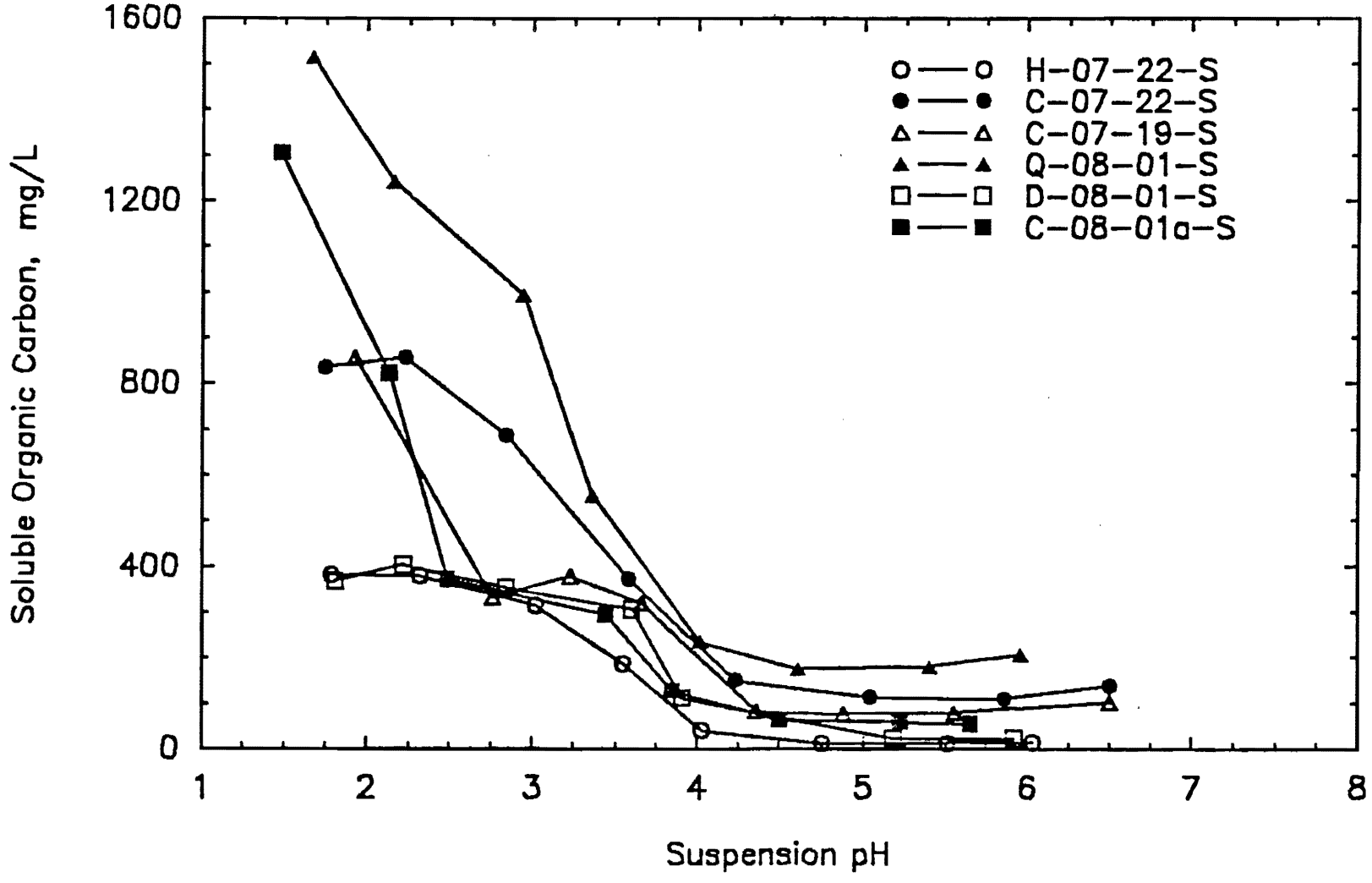

FIGURE 23. Soluble Organic Carbon Concentration as a Function of Suspension pH for Alum Sludges Acidified with Sulfuric Acid (Saunders 1989)

TABLE 30

Soluble Organic Carbon and Humic Acid Concentrations in Sludge Extracts (Saunders 1989)

Sludge Extract	SOC mg/L	Humic Substance-Carbon mg/L	Humic Substance-Carbon % of SOC
C-07-19-S	1,186	930	78
C-07-22-S	981	380	38
C-08-01a-S	779	425	55
H-07-22-S	353	160	46
Q-08-01-S	1,792	1,140	63
D-08-01-S	326	170	52

Priority Pollutant Contaminants. A GC/MS scan of unconditioned raw alum sludge samples, recovered coagulants and commercial coagulants was performed for the priority pollutants listed in Appendix A. A total of six specific purgeable organic compounds were detected in one or more of the unconditioned sludge samples and three acid/neutral extractables were detected in one unconditioned sludge, as presented in Table 31. Chloroform was found in all unconditioned sludge samples at concentrations of 15.5 to 518.6 μg/L and was the most prevalent compound detected in all unconditioned sludge samples. The concentration of 1,1,1-trichloroethane in sample D-08-01 was 160.3 μg/L and was the next most prevalent compound in any unconditioned sludge sample. The remaining compounds detected were pyrene, anthracene, 1,4-dichlorophenol, benzene, toluene, 1,2-dichloroethane and chlorobenzene at concentrations ranging from 1.3 to 17.7 μg/L. The above data indicate that few priority pollutants were contained in the unconditioned sludge samples. Furthermore, those detected in concentrations above 20 μg/L were volatile organic compounds.

Examination of the two commercial coagulants and acid extracted coagulants (C-07-19, H-07-22, D-08-01 and Q-08-01) demonstrated that no detectable priority

TABLE 31

Organic Priority Pollutants Contained in Raw Sludges from Plants in Metropolitan Atlanta, GA (Saunders 1989)

Compound	C-07-22		H-07-22		D-08-01		Q-08-01	
	µg/L	mg/kg SS	µg/L	mg/kg SS	µg/L	mg/kg SS	µg/L	mg/kg SS
PURGEABLES								
Benzene	5.2	0.05	5.5	0.63	-	-	-	-
Toluene	1.3	0.012	-	-	-	-	-	-
Chlorobenzene	-	-	-	-	-	-	1.8	0.018
1,2-Dichloroethane	-	-	-	-	-	-	2.2	0.023
1,1,1-Trichloroethane	-	-	-	-	160.3	14.8	-	-
Chloroform	15.5	0.15	325.0	37.5	309.9	28.6	518.6	5.32
NEUTRAL EXTRACTABLES								
Anthracene	-	-	-	-	-	-	10.2	0.105
Pyrene	-	-	-	-	-	-	17.7	0.18
ACID EXTRACTABLES								
2,4-Dichlorophenol	-	-	-	-	-	-	6.1	0.063

NOTE: The base-extractable priority pollutants, 3,3'-dichlorobenzidine and benzidine, were not examined.

pollutants were contained in these samples. Although the fate of the priority pollutants found in the sludge samples is unclear, the acid extraction procedure did not result in their separation from the sludge solids; resulted in their discharge to the atmosphere; or, resulted in their destruction.

Projected Contaminant Concentrations in Coagulated Water. To determine the potential for contamination of coagulated water by the use of recovered coagulants, metal concentrations were projected for the point of coagulant addition on the basis of an aluminum dose of 1 mg/L. That is, the maximum contribution of metal from the recovered coagulants was determined by normalization to a coagulant dose equal to 1 mg/L as Al. A dose of 1 mg/L as Al, equivalent to 11.1 mg/L as alum, was used as a reference dose, with actual doses in the industry ranging from about 0.03 mg/L as Al to 15 mg/L as Al. The projected metal concentration does not take into consideration the removal of any constituent during the coagulation process. Therefore, these concentrations are the maximum concentrations attributable to the coagulants, but could be considerable lower in the finished water.

Data on the projected metal concentrations at the point of coagulant addition attributable to the recovered coagulants were presented earlier in Table 32 for the Durham coagulant. Comparison of projected metal concentrations to primary MCLs indicated that all controlled metals well within the limits placed on finished drinking waters. Iron and manganese concentrations were high compared to secondary MCLs previously established. The projected iron concentration was 146 μg/L compared to the secondary MCL of 300 μg/L, while the manganese concentration of 127 μg/L exceeded the secondary MCL of 50 μg/L. It must be recognized, however, that these concentrations are projected from the concentration contained in the recovered coagulant. Iron concentrations in coagulated water will be lower since it is least soluble in the neutral pH range and will precipitate as solid hydroxide species during the coagulation process. Manganese concentrations at these levels would require further treatment for oxidation and subsequent removal.

The same evaluation was also performed on the data obtained in the Atlanta study by Saunders (1989) and is presented in Table 33. Comparison to MCLs indicates that all projected metal concentrations are well below the limits

TABLE 32

Projected Metals Concentration at the Point of Coagulant Addition Attributable to an Aluminum Dose of 1 mg/L (Bishop et al. 1987)

Metal	Projected[1] Metal Concentration mg/L	MCL[2] µg/L
Al	--	--
Ag	0	50
As	0.050	--
Ba	0.100	1,000
Ca	1.400	--
Cd	0	10
Cr	0.300	50
Cu	0.300	1,000[3]
Fe	146	300[3]
Hg	0	2
K	3.00	--
Mg	2.70	--
Mn	127	50[3]
Na	3.30	--
Ni	0.03	--
Pb	0.02	50
Se	0	--
Si	4.20	--
Sn	--	--
Zn	0.90	5,000[3]

[1]Projected at the point of coagulant addition prior to removal during coagulation processes
[2]Maximum Contaminant Levels (USPHS 1962)
[3]Secondary MCL values

TABLE 33

Projected Metals Concentration at the Point of Coagulant Addition Attirbutable to Recovered and Commercial Coagulant Addition at a Dose of 1 mg/L as Aluminum (Saunders 1989)

		Chattahoochee					Candler		
Metal	MCL*	C-Alum	C-07-19	C-07-22	C-08-01	H-07-22	D-Alum	D-08-01	Q-08-01
Ag, ng/L	50,000	4.2	0.95	0.9	0.9	4.9	3.6	2.0	1.1
Ba, ng/L	1,000,000	-	-	-	-	-	-	-	-
Cd, ng/L	10,000	0.4	1.1	8.0	14.1	6.9	5.4	29.6	9.4
Cr, ng/L	50,000	1,285	1,463	1,368	2,225	835	727	676	1,196
Hg, ng/L	2,000	0.08	0.2	1.1	0.2	3.5	0.06	2.5	0.3
Pb, ng/L	50,000	299	695	2,160	1,033	763	182	871	597
Cu, ng/L	1,000,000**	91.2	879	2,160	1,900	780	50.9	955	1,706
Fe, mg/L	0.3**	0.03	1.0	1.0	0.4	0.27	0.04	0.17	0.64
Mn, ng/L	50,000**	153	105,260	116,000	79,167	53,922	129	33,636	43,890
Zn, ng/L	5,000,000**	141	2,790	3,760	3,750	1,569	154	909	1,611
Ni, ng/L	-	723.1	-	-	-	-	845	-	-
Sn, ng/L	-	2,570	-	-	-	-	2,816	-	-

*MCL = primary maximum contaminant level MCL for finished drinking water, unless noted otherwise
**Secondary MCL values

NOTE: ng/L = 10^{-3} µg/L = 10^{-6} mg/L

placed on finished drinking waters. As with the Durham coagulant, projected iron and manganese concentrations exceeded the secondary MCL limits. Projected iron concentrations ranged from 170 to 1000 μg/L, while projected manganese concentrations ranged from 44 to 116 μg/L. Projected concentrations of chromium and zinc were also markedly high, but were well below the standards. Comparison to commercial alum products used at the Chattahoochee and Candler plants indicated that chromium concentrations were similar in both commercial and recovered coagulants. Projected nickel and tin concentrations attributable to commercial products were 800 ng/L and 2600 ng/L, respectively, while these constituents were below detection limits in all recovered coagulants studied.

SOC and humic acid concentrations from the Atlanta study (Saunders 1989) were projected to the point of coagulant addition as described above for the inorganic contaminants based on an aluminum dose of 1 mg/L. As presented in Table 34, projected SOC concentrations ranged from 0.35 mg/L to 1.0 mg/L, while projected humic substances ranged from 0.16 mg/L to 0.63 mg/L. These organic carbon concentrations would indicate the potential for increased THM precursor concentrations and may also have a dramatic impact on the utility of the recovered coagulants. However, it should be stressed that these concentrations do not take into account any removal of SOC during the coagulation process. Organic priority pollutant concentrations were not projected to coagulated waters since these compounds were not found at detectable concentrations in the recovered or commercial coagulants.

Treatment of Residual Sludge Solids Following Acidic Extraction

Sludges produced from the coagulation of raw water color and turbidity with alum are bulky, gelatinous suspensions containing aluminum oxides and hydroxides, inorganic particles such as clay, color and colloids, and organic matter found in the raw water (Albrecht 1972). The sludge is generally difficult to dewater and posses a formidable task to the industry. Treatment of alum sludge with acid causes a reduction in both the volume of solids to be handled and the dry mass of solids for disposal.

TABLE 34

Projected SOC and Humic Acid Concentrations at the Point of Coagulant Addition for a Recovered Coagulant Dose of 1 mg/L as Aluminum (Saunders 1989)

Sludge Extract	SOC mg/L	Humic Substance-Carbon mg/L
C-07-19-S	0.62	0.49
C-07-22-S	0.78	0.30
C-08-01a-S	0.62	0.35
H-07-22-S	0.35	0.16
Q-08-01-S	1.0	0.63
D-08-01-S	0.59	0.31

Sludge Volume Reduction

Acidification of aluminum hydroxide sludges results in considerable reductions in residual sludge volume. Acidic extraction dissolves the gelatinous aluminum hydroxide solids and released the bound water associated with the sludge. The volume of the remaining solids is thereby reduced, decreasing the volume of sludge for disposal. As demonstrated by data summarized in Table 35, the resulting reductions in the volume of solids to be handled ranged from 41 to 92.5 percent.

Cornwell and Susan (1979) studied the relationship between sludge volume reduction and aluminum dissolution. Results presented in Figure 24 indicate the dependency of volumetric reduction on aluminum dissolution, with an increasing volume reduction as aluminum dissolution increased. This is an expected result since solid aluminum hydroxide typically is a major portion of the sludge solids present.

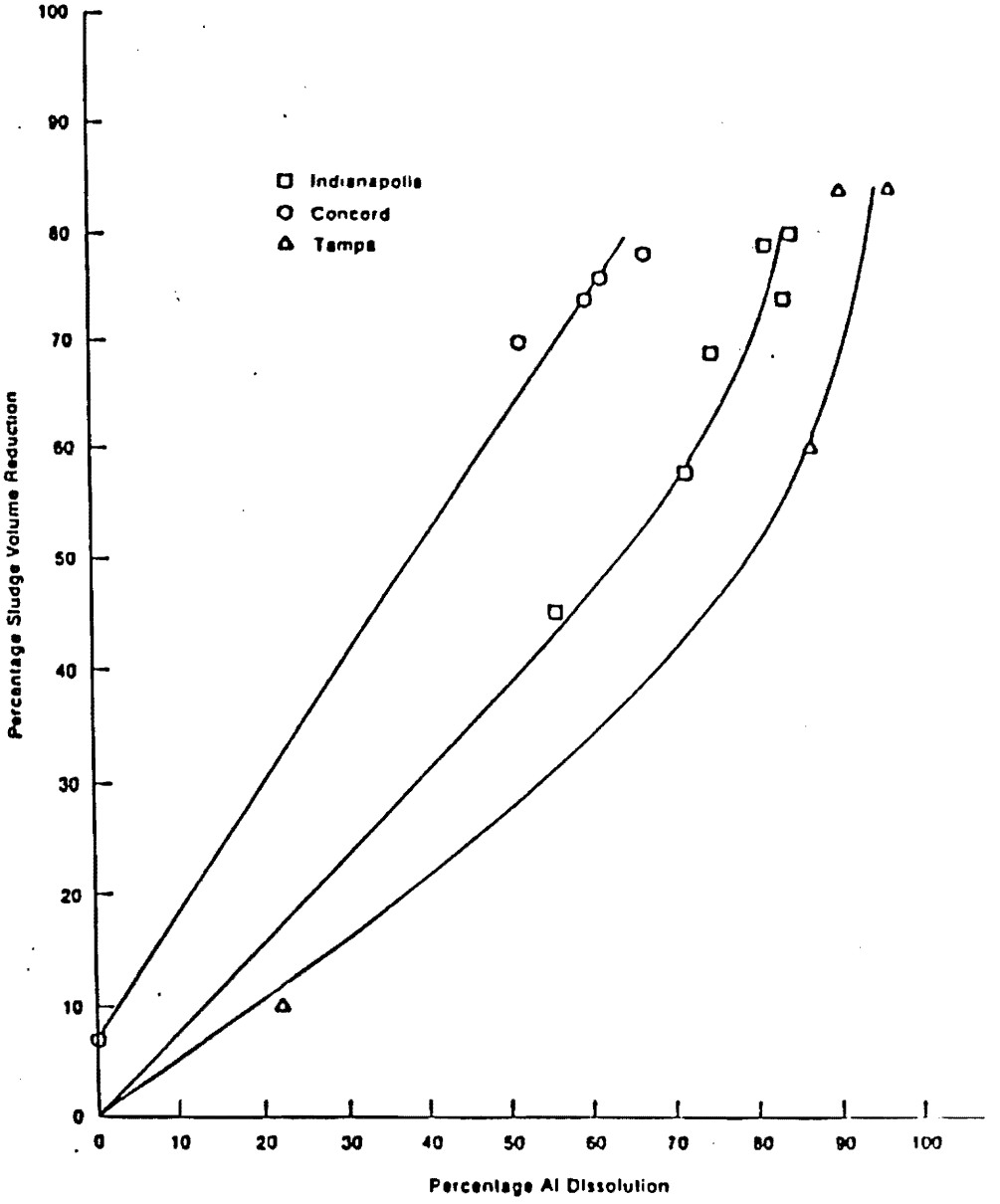

FIGURE 24. Relationship Between Sludge Volume Reduction and Aluminum Dissolution for Three Sludges (Cornwell and Susan 1979)

TABLE 35

Sludge Volume Reduction Following Acidification and Supernatant Recovery (Webster 1969 and White and White 1984)

% Volume Reduction	pH	Raw Sludge Solids %
Webster 1969		
80	3.77	-
82	3.65	-
88.5	3.50	-
90	3.21	-
91	2.98	-
92.5	2.64	-
White and White 1984		
41	-	10.6
54	-	9.6
50	-	3.8
36	-	4.5
36	-	2.5
36	-	3.6
88	-	1.6

Saunders (1989) conducted a study into the thickening properties of unconditioned, acid conditioned, polymer conditioned and lime conditioned slurries. Representative data for one of the sludge samples (H-09-10d) are presented in Table 36. The initial solids concentration of the sludge was 23.5 g/L at a pH value of 7.2. Settled sludge volume for the unconditioned and acid conditioned sludge (pH = 2.0) approached 45 percent of the initial volume in about 2.5 hours, indicating a concentration of sludge solids by a factor of about 2.2. Anionic polymer conditioning (20 ppm) of the acidified sludge suspension resulted in an ultimate settled sludge volume equal to 29 percent of the total, a further decrease in sludge volume.

An acidified, polymer-conditioned sludge sample was then decanted by 45 percent and used for lime conditioning studies. Lime conditioning to a pH value of 9 resulted in a residual sludge volume of only 9.2 percent,

TABLE 36

Thickening Characteristics of Unconditioned, Acid Conditioned, Polymer Conditioned, and Lime Conditioned Sludge Suspensions (Saunders 1989)

Conditioning	Settled Sludge Volume %	Settling Time hr	Interfacial Settling Velocity ft/hr
1. Unconditioned.....	45.4	2.3	0.19
2. Acid addition (pH=2).....	45.3	2.5	0.23
3. Acid addition (pH=2); polymer addition (20 ppm).....	29	12	--
4. Acid addition (pH=2); polymer addition; decant 45% of clarified liquid:			
a. Lime addition (pH=9).....	9.2	23	0.0045
b. Lime addition (pH=10).....	2.6	2.5	0.0096
c. Lime addition (pH=11).....	20.3	2.3	0.14
d. Lime addition (pH=12).....	43.8 24.6	1.0 2.0	0.27 0.24

corresponding to a 10.8-fold increase in solids concentration. Further lime addition up to a pH value of 12 resulted in a dramatic increase in interfacial settling velocity. However, the settled sludge volume also increased due to the addition of lime and the precipitation of insoluble calcium aluminates, which contribute to the sludge solids. Therefore it was concluded that the acidified sludges could be effectively thickened by gravity settling following polymer addition and that lime addition to polymer conditioned sludge could be used to further consolidate the sludge solids prior to dewatering.

In full-scale testing at Durham, NC, sludge volume was reduced from 265 m^3 to 83 m^3 (69 percent) in test 1 and 292 m^3 to 136 m^3 (53 percent) in test 2 (Bishop et al. 1987). An analysis of required sludge drying bed area found that use of an alum recovery process for sludge volume reduction would result in a decrease in required drying bed area by a factor of approximately 2.7.

Solids Reduction and Concentration

Reduction of sludge solids is the result of dissolution of aluminum and other soluble constituents in the sludge during acid extraction. Westerhoff (1973) reported a 13 percent solids reduction in pilot-scale testing at Sturgeon Point. Cornwell and Susan (1979) investigated the dissolvable solids at a pH value of 1, as presented earlier in Table 21. The dissolvable solids ranged from 9 percent to 86 percent of the total suspended solids, showing the potential for solids reduction at reduced pH values. Cornwell and Susan (1979) concluded from data in Figure 25 that the reduction in suspended solids was directly proportional to percent aluminum dissolution. Although data presented in Figure 25 show an increase in suspended solids reduction as aluminum dissolution increased, the lack of data in the range of 10 to 50 percent aluminum dissolution and the concentration of data points at a higher percent aluminum dissolution does not necessarily support the direct proportionality indicated in the figure. It was concluded that aluminum hydroxide sludges resulting from large alum doses on raw water which is high in color and low in turbidity were found to experience the largest solids reductions since aluminum hydroxide made up such a large portion of the sludge solids.

In full-scale testing, Bishop et al. (1987) observed 53 and 42 percent solids

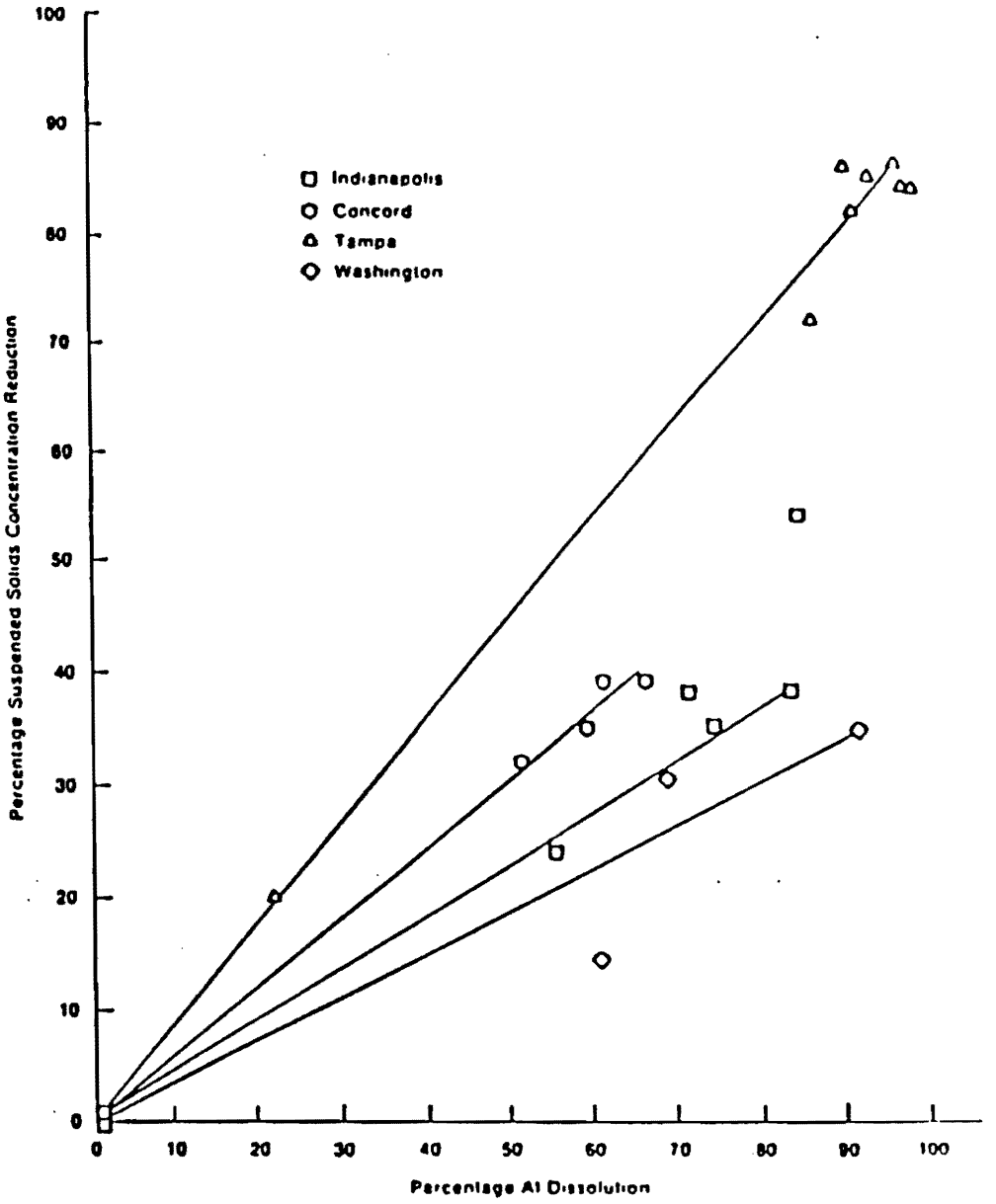

FIGURE 25. Relationship Between Suspended Solids Reduction and Aluminum Dissolution for Acidified Alum Sludges (Cornwell and Susan 1979)

reductions as the result of acidification. This resulted in a reduction of sludge solids being applied to sand drying beds from 6350 kg and 9757 kg to 2994 kg and 5706 kg for tests 1 and 2, respectively. As discussed previously, the required drying bed area was reduced by a factor of about 2.7.

Dewaterability of Residual Sludge Solids

The dewaterability of residual sludge solids has been investigated using many methods of testing and in some cases compared to unconditioned sludge suspensions. Capillary suction time, specific resistance, filter press operation, sand drying beds, pressure flotation and the freeze-thaw process have all been used as measures of the dewaterability of acidified alum sludge suspensions. Lime and polymer conditioning of these suspensions have also been investigated to determine their impact on thickening and dewatering properties.

Specific Resistance. Specific resistance is often used as a quantitative measure of the filterability of a sludge. King et al. (1975) reported a sharp reduction in the specific resistance of acidified alum sludges in the range of 60 to 80 percent aluminum recovery. As presented in Figure 26, specific resistance increased dramatically as aluminum recovery increased beyond this point, indicating a deterioration in filterability of the sludge with a further increase in recovery. Albrecht (1972) reported specific resistance of alum sludges conditioned with sulfuric acid, lime, anionic polymer, cationic polymer, or a combination thereof, as presented in Table 37. It was noted that lesser amounts of conditioning chemicals were required for adequate conditioning of acidified sludge and higher cake solids concentrations were obtained. The anionic polymer was ineffective on the acidified sludge, while conditioning with a cationic polymer proved advantageous. Isaac and Vahidi (1961) reported that when the pH of alum sludge was reduced from about 7 to 5 with the addition of sulfuric acid, the specific resistance of the sludge was reduced more than two-fold.

Sand Drying Beds. Bishop et al. (1987) reported two full-scale tests using sand drying beds for dewatering acidified sludge solids. Test 1 used an anionic polymer as pretreatment before application to the beds. This resulted

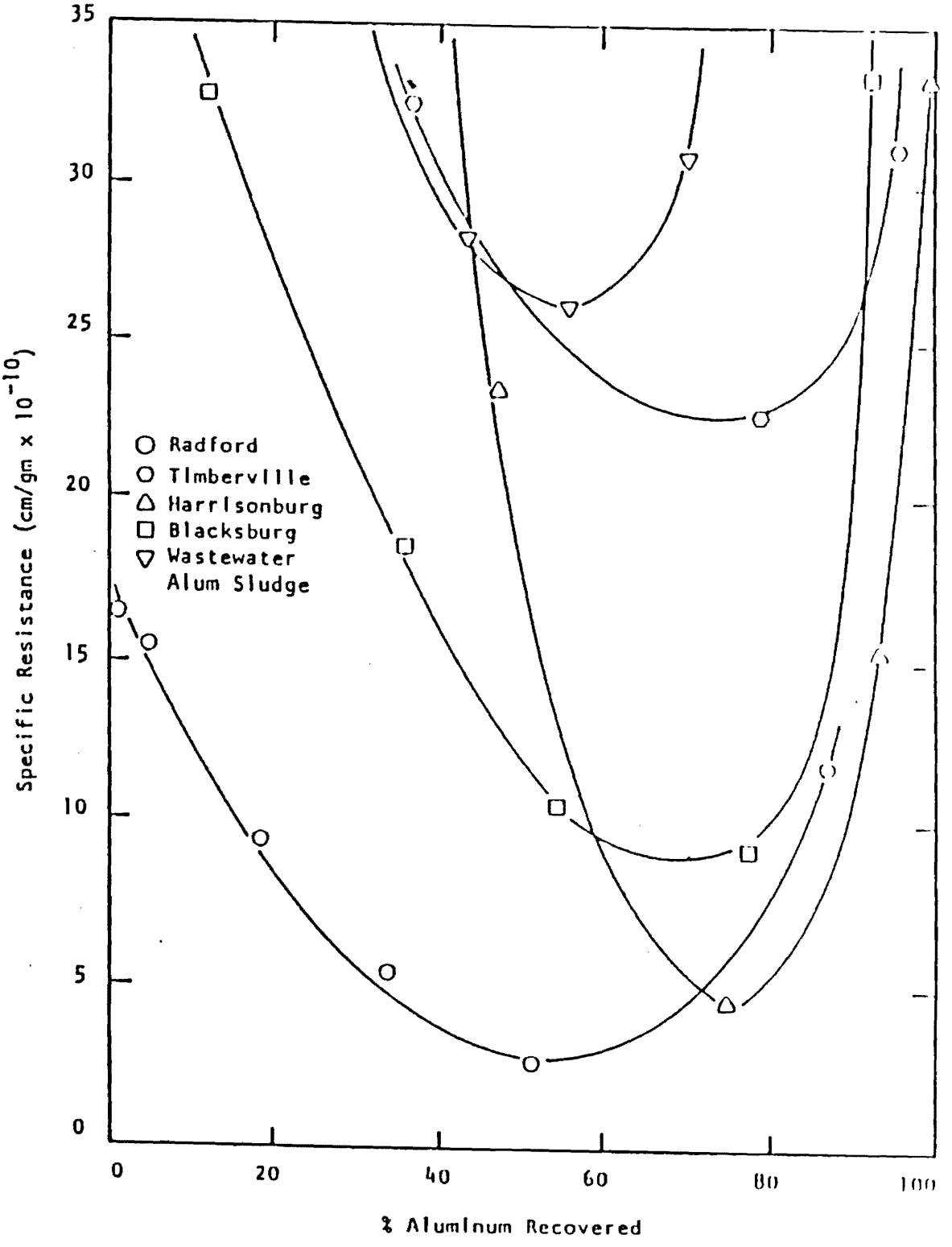

FIGURE 26. Relationship Between Aluminum Dissolution and Specific Resistance of Acidified Alum Sludges (King et al. 1975)

TABLE 37

Effect of Acid, Lime and Polymer Conditioning on Cake Solids Concentration and Specific Resistance (Albrecht 1972)

Sludge Solids mg/L	Vacuum in. Hg	Acidified		Conditioning Agent* per cent by weight		Cake Solids percent	Specific Resistance Tm/kg
		Yes	No	Lime	Polymer		
7,700	18		X		0.075A	15.6	5.1
7,700	18		X	15.0	0.075A	21.8	2.45
7,700	18		X	26.0	0.15A	25.0	3.92
7,700	18		X	20.0	0.05A	24.4	2.75
7,700	27		X	20.0	0.1A	22.0	2.65
7,700	20		X	20.0	0.1A	20.0	1.47
7,700	12		X	20.0	0.1A	18.8	0.726
11,000	18	X			0.075C	28.6	0.255
11,000	18	X			0.15C	36.3	0.128
11,000	18	X				28.0	0.157

*A - Anionic polyelectrolytic chemical aid; C - Cationic polyelectrolytic chemical aid

in a decrease in capillary suction time (CST) from 288 to 130 sec, while 36 percent of the volume applied to the bed was recovered by underdrains and decant piping. Test 2 differed from Test 1 only in that a cationic polymer was used. This resulted in a reduction in cst to less than 20 seconds, while 53 percent of the volume applied was recovered. The cationic polymer appeared to be superior to the anionic polymer due to the improved dewatering properties of sludge from test 2. It was concluded that use of coagulant recovery resulted in a reduction of sludge volume and solids to be dewatered and decreased the required drying bed area by 62 percent. The resultant solids concentration on the sand drying bed ranged from 14.3 to 18.5 percent.

Pressure Flotation. Albrecht (1972) used pressure flotation as a means of dewatering an acidified alum sludge. This method was chosen due to gas bubbles observed in the acidified sludge interfering with gravity settling. It was thought that this phenomena would enhance the performance of the pressure flotation process. As presented in Table 38, acidified alum sludge, treated with cationic polymer dewatered much better than raw alum sludge. The cationic polymer proved to be superior to the anionic polymer, in agreement with the results of Bishop et al. (1987).

TABLE 38

Flotation Thickening Characteristics of Raw and Acidified Sludge Samples (Albrecht 1972)

Sludge	Chemical[1] Aid, mg/L	Initial Solids Concentration Percent	Final Solids[2] Concentration Percent	Recycle Rate Percent	Rise Rate ft/min
Raw	8-A	0.77	1.6	148	3.9
Acidified	None	0.40	3.7	74	4.3
Acidified	8-C	0.40	4.0	74	>7.0

[1]A - Anionic polyelectrolytic chemical aid
C - Cationic polyelectrolytic chemical aid
[2]Average concentration based on change of sludge volume

Freeze-Thaw. Webster (1969) used freeze-thaw techniques for the consolidation and dewatering of acidified alum sludges on both a laboratory and pilot scale level. A 5-L volume of alum sludge was removed from a pilot plant sedimentation basin to be tested for alum recovery and sludge solids concentration. Following acidification and settling, the sludge volume was reduced by 90 percent, i.e., 90 percent of the volume was recovered as a coagulant. After the freeze thaw process, the residual sludge volume was further reduced by 86 percent. The overall result was a sludge that had been reduced in volume by greater than 98 percent through use of acidification and a freeze-thaw process.

The results obtained from the pilot plant were extrapolated to predict the performance of a full-scale plant treating 118,000 m^3/d of water for potable use. Acidic coagulant recovery and freeze-thawing were utilized to treat residual sludge solids. A 378.5 m^3 volume of 0.5 percent sludge solids would be pumped once daily to the acidification plant where the pH would be adjusted to 2.5 with concentrated sulfuric acid. Following 12 hours of slow stirring, about 288 m^3 (76 percent) of supernatant liquid would be recovered and sent to alum storage tanks while the remaining sludge would be pumped to separator tanks where 71 m^3 (17 percent) of supernatant liquid would be recovered. The remaining 20 m^3 of sludge would be pumped to the refrigeration house where 15.5 m^3 of recovered alum would be generated leaving the remaining 4.5 m^3 to be deposited on waste land. This residual sludge volume would be less than two percent of the initial raw sludge volume with the remainder of the liquid volume being recovered for use as a coagulant.

ALUMINUM RECOVERY BY ALKALINE EXTRACTION

Experimental investigations of the recovery of aluminum from alum sludges using alkaline extraction has been limited, compared to acidic extraction. Sodium hydroxide (NaOH) and lime (CaO) have been used as conditioning agents to raise the pH of sludge suspensions and dissolve sludge aluminum, present predominantly in the form of $Al(OH)_3 \cdot 3H_2O$. Referring to the aluminum pC-pH diagram presented in Figure 1, in the alkaline pH range solubilized aluminum would be present predominantly as $Al(OH)_4^{1-}$. Soluble aluminum concentrations

can range from 27 mg/L at a pH of about 10 to about 2700 mg/L at a pH value of 12, indicating the potential for alkaline recovery.

The process of alkaline aluminum recovery is similar to that for acidic recovery, as presented earlier in Figure 6. Thickened alum sludge is contacted with alkali in a rapid mix unit and transferred to a separator. Supernatant liquid is then recovered for reuse as a coagulant in water treatment, while sludge from the underflow of the separator is dewatered and the residual sludge solids are disposed of.

Operational Variables Affecting Alkaline Extraction

A number of parameters have been identified as having significant impact on the feasibility of alkali aluminum recovery. These parameters include: stoichiometry of alkali additions; pH of alkali-conditioned suspensions; and, kinetics of alkaline extraction. These parameters will impact the concentration of aluminum in recovered coagulants and its quality for reuse as a coagulant in the water treatment industry.

Stoichiometric Evaluation of Experimental Alkali Additions

Sodium hydroxide and lime have been investigated for use in alkali extraction of aluminum from alum sludges produced in the treatment of drinking water. The reactions involved in alkali extraction of aluminum using sodium hydroxide and calcium hydroxide can be summarized as follows:

$$Al(OH)_3 \cdot 3H_2O + NaOH = NaAl(OH)_4 \quad (3)$$

$$2Al(OH)_3 + Ca(OH)_2 = CaAl_2(OH)_8 \quad (4)$$

The stoichiometric demand for NaOH is 1 mole/mole aluminum or 1.48 kg/kg Al (equation 3), while the stoichiometric demand for $Ca(OH)_2$ is 0.5 moles/mole Al or 1.37 kg/kg aluminum (equation 4). These reactions are written assuming complete dissolution of solid $Al(OH)_3 \cdot 3H_2O$ to soluble $Al(OH)_4^-$. The possibility of the formation of other negatively-charged aluminum species does exist and can have a significant impact on the stoichiometry of aluminum dissolution. King et al. (1975) showed that the reactions involving NaOH and

$Ca(OH)_2$ did not follow stoichiometric predictions (Figure 7). It was concluded that in the case of recovery using lime, low $Ca(OH)_2$ concentrations and the limited solubility of calcium aluminates formed in the reaction were the main contributors to the lack of success.

The effects of calcium ions on the dissolution of aluminum was investigated by Holmes (1982). Sodium hydroxide was first used as a conditioning agent for pH adjustment and aluminum dissolution. Calcium ions were then added in the form of $CaCl_2$ to determine their impact on soluble aluminum concentrations. It was concluded that the addition of calcium to the suspensions caused the formation of sparingly soluble calcium aluminates, significantly decreasing the soluble aluminum present in the extracts and increasing the volume of residual sludge solids due to calcium aluminate precipitation. These results help explain the problems encountered with the use of lime for alkaline extraction and will be referred to later.

Chemical additions greater than those predicted stoichiometrically have been required to acheive adequate recovery in most investigations using both lime and sodium hydroxide. Masschelein et al. (1985) reported on an extensive study of alkaline extraction of aluminum from raw alum sludges. A sodium hydroxide requirement of 1.75 mole/mole of recovered aluminum (1.48 kg/kg Al recovered) was reported on sludges with suspended solids concentrations ranging from 1.0 to 1.5 percent dry solids. Recovery of up to 80 percent of the total aluminum was realized, with a resultant maximum supernatant aluminum concentration of about 945 mg/L at a pH of 11.6. The requirement for $Ca(OH)_2$ ranged from 7 to 8 moles/mole for the same sludge, with recovery of less than 25 percent of the total aluminum. The maximum supernatant aluminum concentration using lime was about 270 mg/L at a pH of 11.4.

Wang and Yang (1975) presented data obtained using sodium hydroxide and lime for alum recovery on a sludge containing 156 mg Al/g dry sludge solids. This was equivalent to 42.9 mg Al/g wet sludge solids. Varying amounts of a saturated lime solution were mixed with approximately 5 g of wet sludge, resulting in a maximum of only 16 mg of aluminum being recovered (7.4 percent of the total) as indicated in Table 39. Use of sodium hydroxide proved to be more successful, with as much as 100 percent of the total aluminum being

TABLE 39

Aluminum Recovery from Alum Sludges Using Lime [$Ca(OH)_2$] for pH Adjustment (Wang and Yang 1961)

Mass Wet Sludge (g)	Total Sludge Al, mg	Extractant[1]	Final pH	Recovered Aluminum		
				mg/L	mg	% of total
5.013	215	100 mL distilled water	7.71	<10	<1	<0.5
5.160	221	40 mL saturated lime 60 mL distilled water	11.70	<10	<1	<0.5
5.507	236	100 mL saturated lime	12.16	30	3	1.3
5.140	221	200 mL saturated lime	12.35	30+	6	2.7
5.189	222	300 mL saturated lime	12.33	20	6	2.7
5.012	215	800 mL saturated lime	12.44	20	16	7.4
5.125	220	5.1 g lime and 800 mL distilled water	12.48	20	16	7.3

[1]Mixed with the indicated mass of wet sludge solids

recovered using NaOH doses of 32-91 kg/kg recovered aluminum, as presented in Table 40. Although 100 percent recovery was achieved, these doses were 22- to 61-fold higher than stoichiometric requirements (i.e., 1.48 g NaOH/g Al).

pH of Alkali Conditioned Sludge Samples

Masschelein et al. (1985) reported using CaO and NaOH for aluminum extraction on sludges of varying solids and aluminum concentrations. Alkaline aluminum recovery was performed to examine the effects of varying solids concentrations and extraction pH. A suspended solids concentration of less than 20 g/L was established for optimum supernatant recovery, since at higher solids concentrations less than 50 percent of the volume could be recovered by gravity settling (Figure 27). After establishing this relationship, sludges containing 10 to 15 g/L of dry solids were reacted with CaO or NaOH to establish optimum pH ranges for each. Results obtained using lime are presented in Figure 28. Aluminum concentration was maximized in the pH range of 11.2 to 11.6 where the recovered concentration was about 270 mg/L as Al (10 mmole/L), approximately 25 percent of the total aluminum present. Above a pH of 11.6, soluble aluminum concentrations decreased dramatically. These results seem to follow those obtained by Holmes (1982) in that as the calcium concentration increased, the soluble aluminum concentration decreased. Use of NaOH proved to be much more successful than lime. As presented in Figure 29, an optimum pH range of 11.4 to 11.8 was established where aluminum recovery was maximized. The resulting recovered aluminum concentration was approximately 950 mg/L (35 mmole/L) and represented about 80 percent of the total aluminum present.

Slechta and Culp (1967) reported 90 percent recovery at pH 12 using NaOH. The 90 percent recovery was based on supernatant liquid recovery assuming complete aluminum dissolution (i.e., 90 percent volume reduction of the sludge). Complete aluminum dissolution, however, was not confirmed. Filtration techniques for supernatant recovery proved to be more efficient than did sedimentation. Using filtration, a maximum of 90 and 35 percent supernatant recovery was possible with NaOH and $Ca(OH)_2$, respectively, while only 75 and 25 percent supernatant were recovered using sedimentation, as presented in Figures 30 and 31.

TABLE 40

Aluminum Recovery from Alum Sludges Using NaOH for pH Adjustment (Wang and Yang 1961)

Mass Sludge (g)	Total Sludge Al, mg	Mass NaOH	Volume of Suspension	Final pH	Recovered Aluminum		NaOH g/g Al Recovered
					mg	% of total	
5.0130	215	0.0[1]	100 mL	7.71	<1	<0.5	0.0
5.0161	215	0.08 g[1]	100 mL	11.46	2.5	1.2	32.0
5.0881	218	0.4 g[1]	100 mL	12.61	5.0	2.3	80.0
5.2249	224	2 g[1]	100 mL	13.28	37.0	16.5	54.1
5.0000	215	4 g	100 mL	13.50	51.0	24	78.4
5.1678	222	10 g	250 mL	13.58	122.5	55	81.6
5.1109	219	20 g	500 mL	13.67	220.0	100	90.9

[1]1-N NaOH solution used in all experiments, but was diluted to 100-mL volume to adequately suspend the sludge and to provide an adequate volume for mixing.

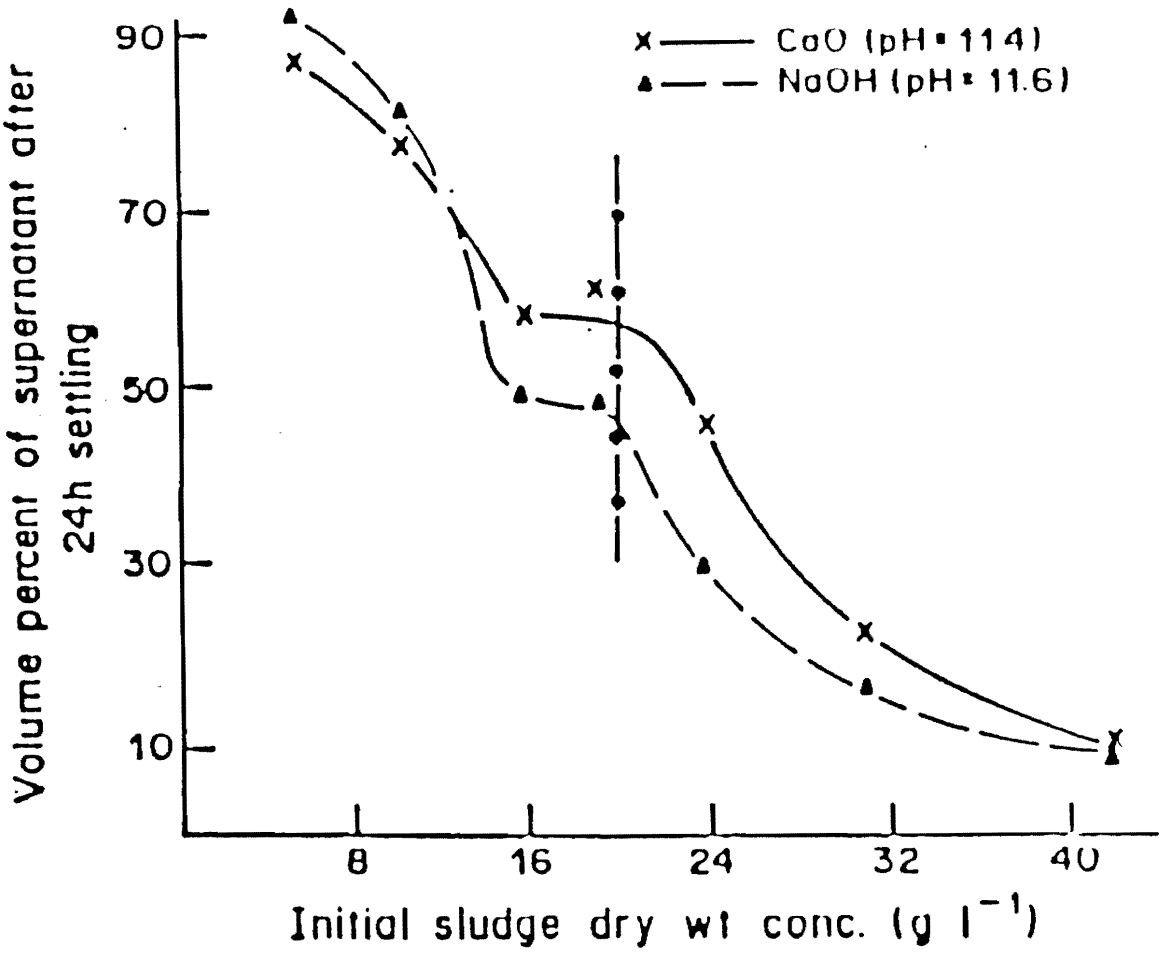

FIGURE 27. Volumetric Recovery of Clarified Supernatant Following CaO and NaOH Conditioning of Alum Sludges (Masschelein et al. 1985)

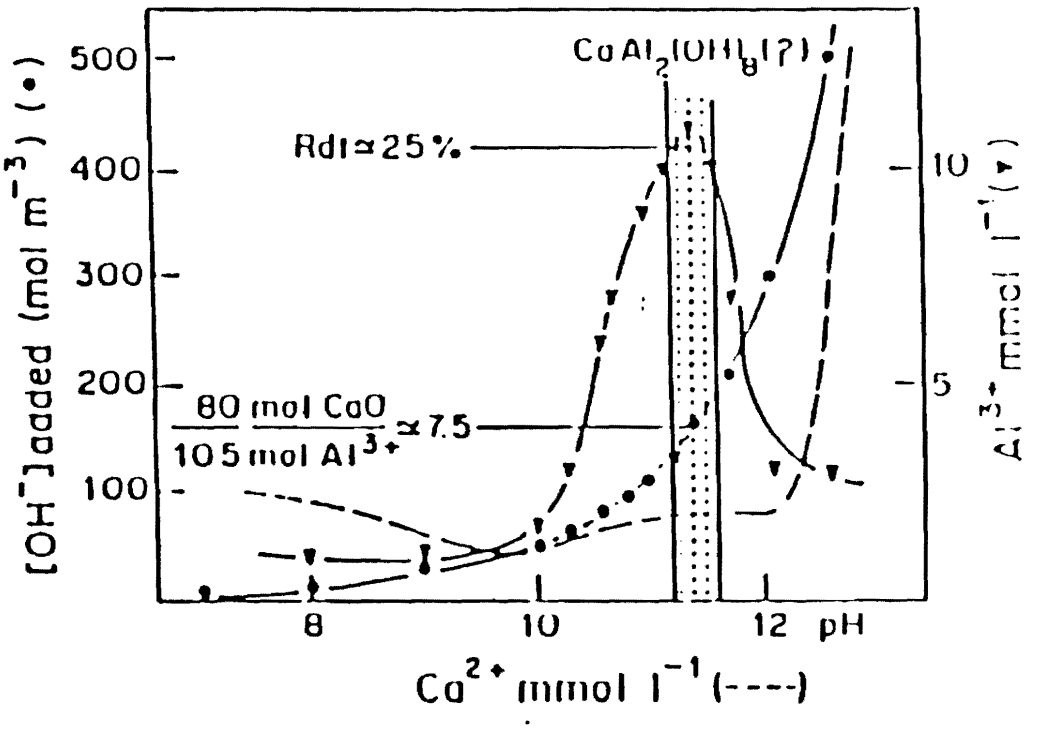

FIGURE 28. Aluminum Dissolution as a Function of Suspension pH Following CaO Addition (Masschelein et al. 1985)

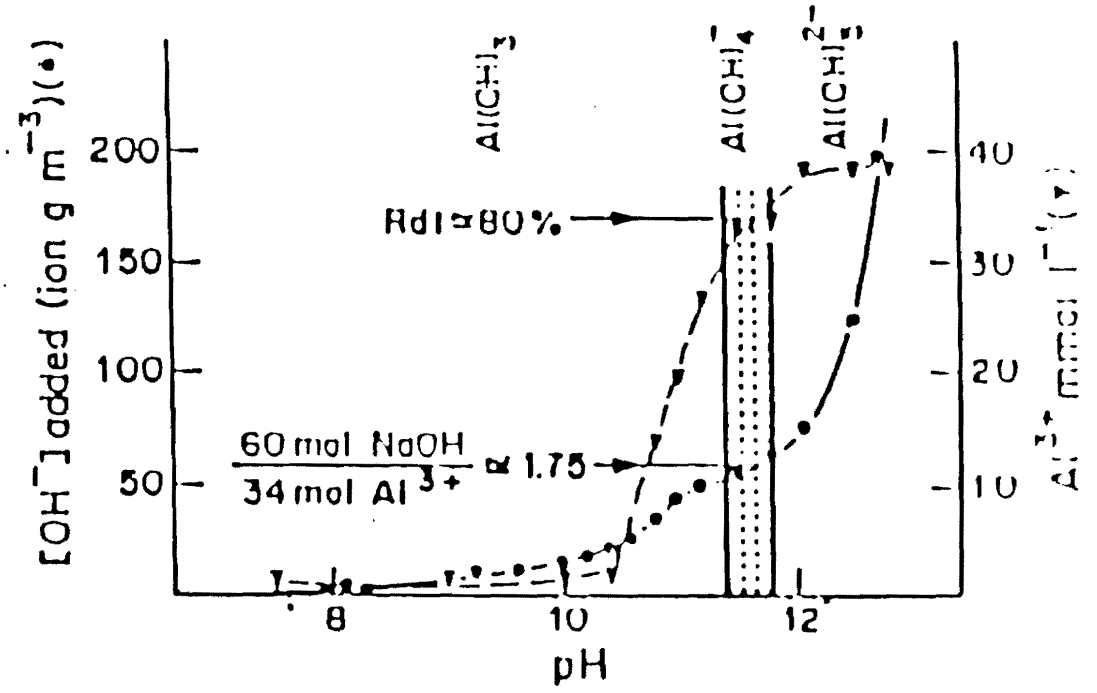

FIGURE 29. Aluminum Dissolution as a Function of Suspension pH Following NaOH Addition (Masschelein et al. 1985)

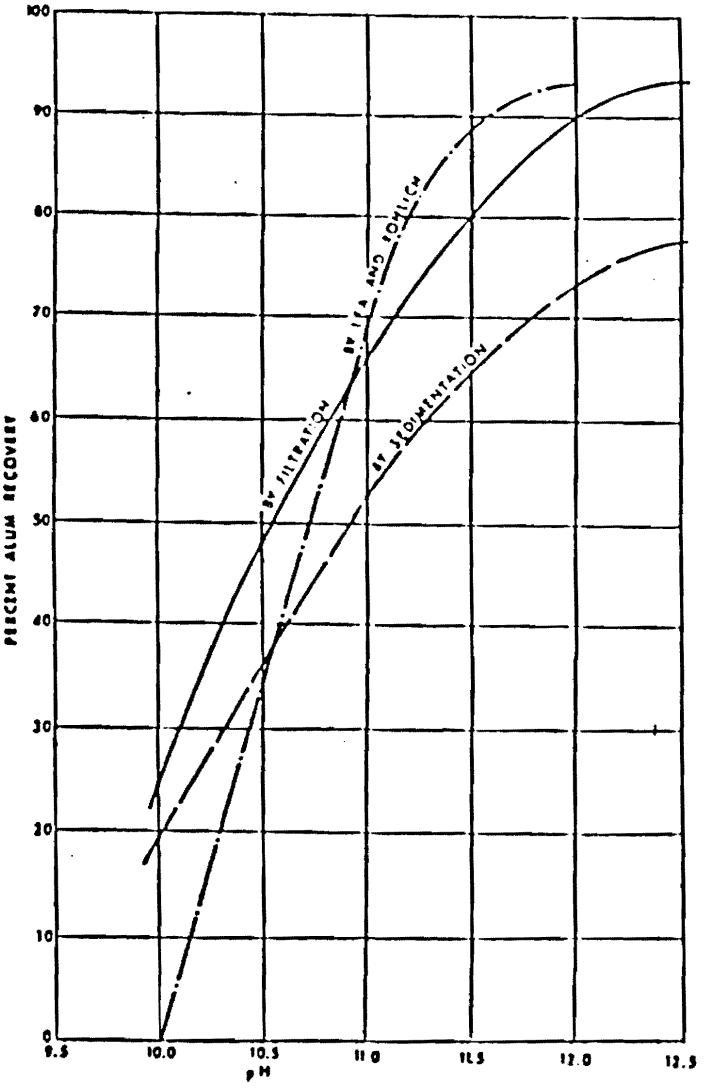

FIGURE 30. Effect of pH on Volumetric Recovery of Clarified Supernatant Liquid Using Sedimentation and Filtration Processes for Solids Separation Following NaOH Addition to Alum Sludges (Slechta and Culp 1967)

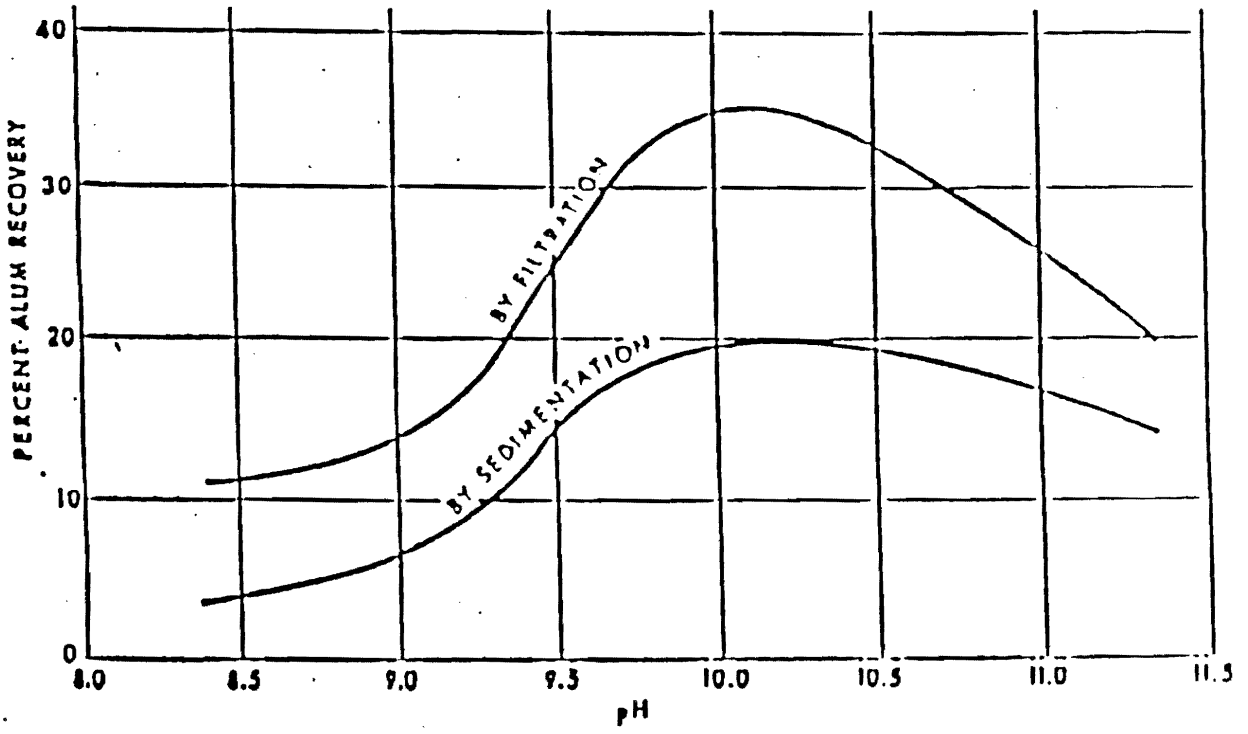

FIGURE 31. Effect of pH on Volumetric Recovery of Clarified Supernatant Liquid Using Sedimentation and Filtration Processes for Solids Separation Following $Ca(OH)_2$ Addition to Alum Sludge (Slechta and Culp 1967)

Wang and Yang (1961) reported coagulant recovery as a function of pH and chemical addition using NaOH and $Ca(OH)_2$. It was concluded that recovery using $Ca(OH)_2$ (Table 39) proved unsatisfactory due to the low $Ca(OH)_2$ concentration in the lime solution and the low solubility of the calcium aluminates formed, agreeing with results obtained by King et al. (1975) and Holmes (1982). Recovery of aluminum as a function of pH using NaOH was presented earlier in Table 40. Extremely high pH values (>13.5) were required to recover about 100 percent of the total aluminum.

Finally, Isaac and Vahidi (1961) concluded that aluminum recovery using NaOH was not satisfactory up to a pH of 11 since only 15 percent recovery was experienced from a sludge containing 0.67 percent aluminum.

Reaction Kinetics of Alkaline Extraction

Wang and Yang (1975) presented aluminum recovery as a function of time using two doses of NaOH, as presented in Table 41. Approximately 5 g of wet sludge was reacted with 250 mL and 500 mL of 1 N NaOH, respectively, for tests A and

TABLE 41

Results of Kinetic Tests on Aluminum Dissolution Following NaOH Addition to Alum Sludge Samples (Wang and Yang 1961)

Time	Recovered Al			
	Test A[1]		Test B[2]	
min	mg/L	mg	mg/L	mg
0	0	0	0	0
5	490	122.5	410	205
10	490	122.5	430	215
30	490	122.5	430	215
60	490	122.5	440	220
960	490	122.5	440	220

[1]NaOH dose: 81 kg/kg recovered Al
[2]NaOH dose: 162 kg/kg recovered Al

B. These doses were equivalent to about 81 kg/kg and 162 kg/kg of aluminum recovered and were about 55- to 109-fold higher than the stoichiometric demand discussed earlier. Five minutes proved to be adequate for recovery of a maximum amount of aluminum, with about 55 and 100 percent of the total aluminum being recovered for tests A and B, respectively. Mixing times of up to 16 hours did not improve recovery significantly beyond that recoverable at 5 minutes. It should be noted that the extreme NaOH doses used in the kinetic tests may have influenced the required mixing time, i.e., at NaOH doses closer to stoichiometric predictions, a larger detention time may be required for adequate recovery.

Quality and Utility of Alkaline Recovered Coagulants

Quality of the products produced in the alkaline extraction of aluminum from alum sludges was based on aluminum content and the presence of organic matter, color and other trace contaminants. Utility of the alkaline extracted coagulants was typically measured using a standard jar test procedure and the coagulated water tested for color, turbidity and other water quality parameters.

Aluminum Content

Masschelein et al. (1985) reported recovered aluminum concentrations as a function of pH, as presented in Figures 29 and 30 using CaO and NaOH, respectively. In the optimum pH range of 11.4 to 11.8 established for NaOH, the aluminum content was found to be approximately 35 mmole/L or 945 mg/L as Al. The optimum recovery range for CaO at a pH of 11.2 - 11.6 resulted in recovered aluminum concentrations of approximately 10 mmole/L or 270 mg/L. Recovered aluminum in the supernatant liquid was also dependent on initial sludge solids concentrations. The percent of total aluminum in supernatant liquid following 24-h settling is presented as a function of solids concentration in Figure 32. The data were obtained at the previously determined optimum pH values of 11.4 and 11.6 for CaO and NaOH extractants, respectively. Recoverable aluminum (percent of total aluminum extracted) in the supernatant liquid dramatically increased as the solids concentration decreased below about 20 g/L. Maximum dissolution appeared to occur at a sludge solids

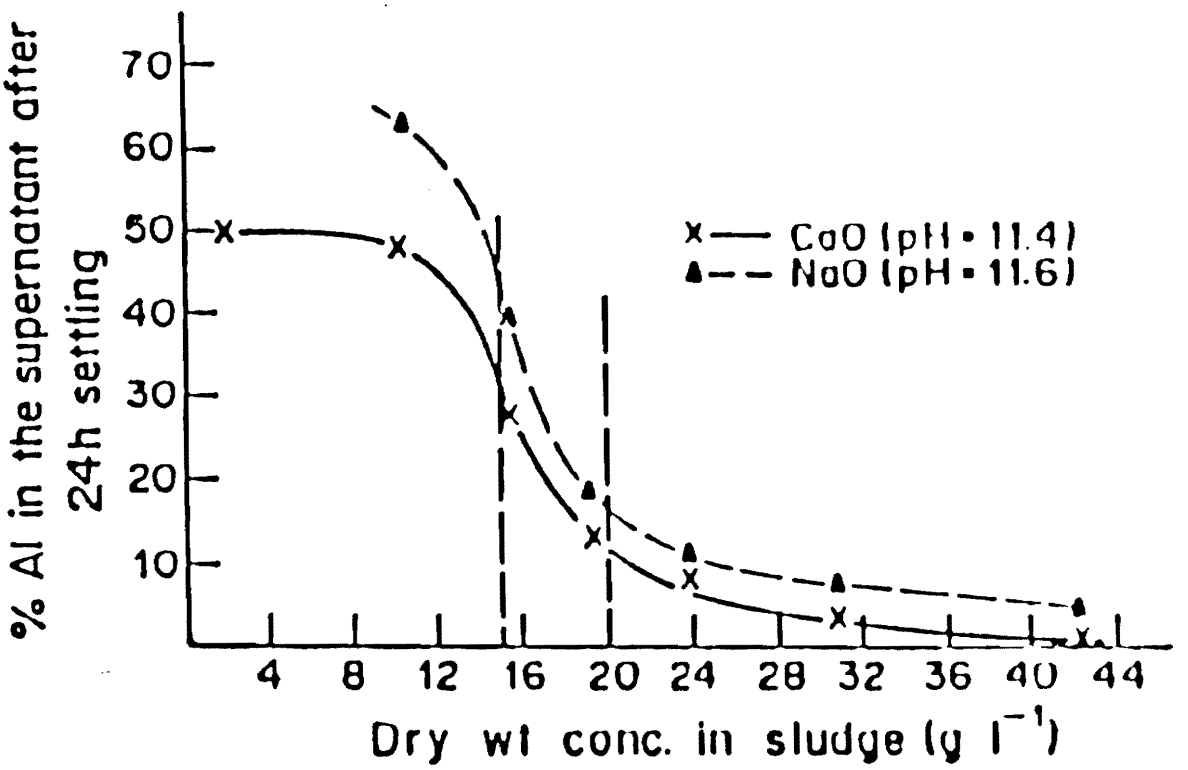

FIGURE 32. Percent Aluminum Recovery as a Function of Initial Sludge Suspended Solids Concentration Following Conditioning of Alum Sludge with CaO and NaOH (Masschelein et al. 1985)

concentration below about 8 g/L as indicated by the plateau of aluminum concentration for both extractants. Resulting maximum aluminum recoveries of about 50 percent and 65 percent were realized for CaO and NaOH extraction, respectively.

Wang and Yang (1961) reported maximum recovered coagulant concentrations for $Ca(OH)_2$ and NaOH to be 30 and 510 mg/L, respectively, in laboratory-scale testing as presented earlier in Tables 39 and 40. These concentrations, however, resulted from treatment of approximately 5 g of wet sludge with volumes of up to 800 mL of extractant. Since large volumes of relatively dilute extractant were added to a small volume of sludge, recovered supernatant aluminum concentrations reported were much lower than what would typically be expected due to the dilute nature of the extractant. Therefore, these numbers are not indicative of typical recovered supernatant aluminum concentrations, however, they can be used to measure total aluminum recovered from the sludge samples. Total aluminum recovered on a mass basis and as percent of total aluminum are also presented in Tables 39 and 40. Sodium hydroxide doses up to 90 g/g of recovered aluminum resulted in recovery of 100 percent of the total. A mass application for calcium hydroxide could not be determined, but only 7.4 percent of the total aluminum was recovered at pH values up to about 12.5 using lime.

Coagulation Effectiveness of Alkaline Recovered Coagulants

Slechta and Culp (1967) reported using alkali-recovered coagulants for the removal of turbidity, although the source of turbidity was not reported. Commercial alum was used in conjunction with recovered supernatant from a NaOH extraction procedure. Varying amounts of commercial alum were added with recovered coagulants to enhance coagulation with the results being compared to coagulation using 100 percent commercial alum. Although the total aluminum dose was nearly twice as high, an equal mixture of commercial alum and recovered aluminum reduced turbidity to the same level as commercial alum (Figure 33). Coagulant recovered using lime was reportedly more effective in removal of turbidity than its NaOH counterpart, although no data were presented. The authors concluded, however, that both methods of coagulant recovery were not economically feasible since such large make-up doses of

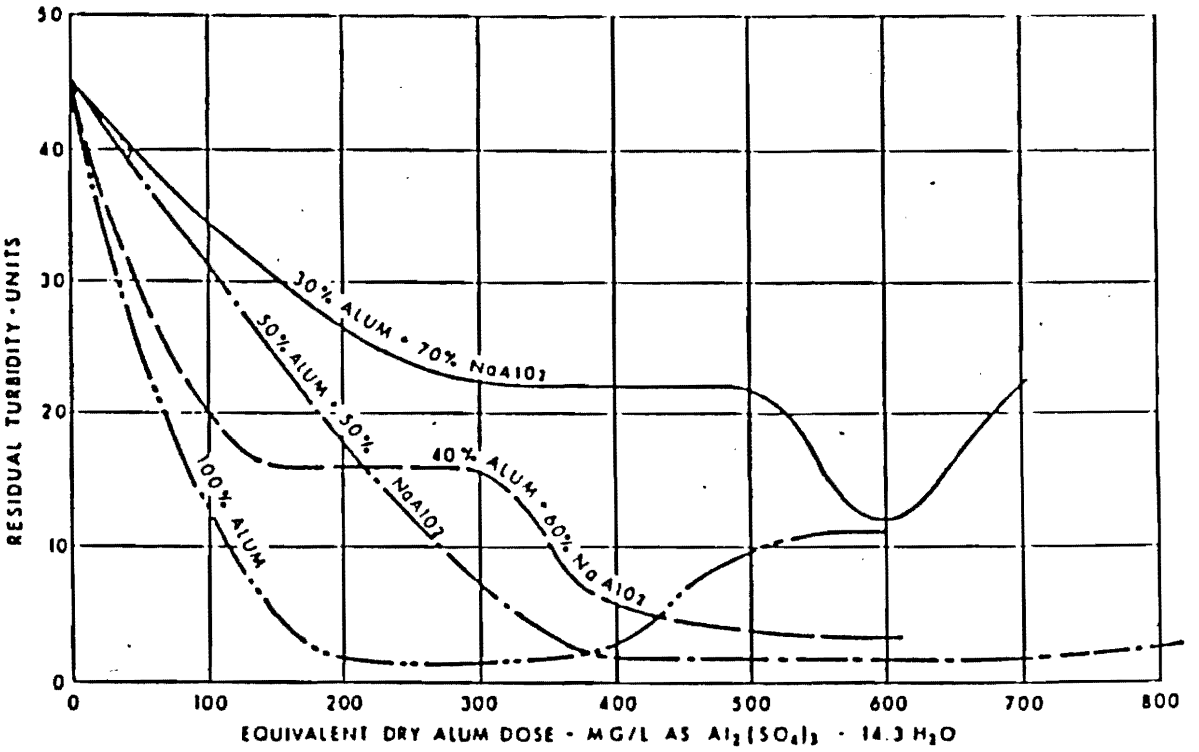

FIGURE 33. Residual Turbidity Following Aluminum Coagulation and Sedimentation in Jar Tests Using Various Combinations of Commercial and NaOH-Recovered Coagulants (Slechta and Culp 1967)

commercial alum were needed to enhance coagulation.

Masschelein et al. (1985) reported the results of coagulation of a natural river water using coagulant recovered with CaO, as presented in Table 42. With up to 50 percent recycle of recovered alum (i.e., 50 percent make-up using commercial polyaluminum chloride), no significant decrease in finished water quality was detected. The pH of the water treated with recovered coagulant was higher than that treated with commercial alum, causing an increase in dissolved aluminum in the finished water. Adjustment of the pH decreased the residual soluble aluminum in the treated water. It was concluded that 25 to 50 percent recycle of coagulant (i.e., 75 to 50 percent make-up with commercial alum) was optimum, causing no significant decrease in finished water quality.

Wang and Yang (1961) reported using aluminum extracted from alum sludges with NaOH for coagulation of raw creek waters. A coagulant dose of 17 mg/L of recovered sodium aluminate was followed by pH adjustment and addition of 3 mg/L of activated carbon and 5 mg/L of chlorine (Cl_2). This finished water

TABLE 42

Settled Water Quality[1] Using Commercial and NaOH-Recovered Coagulants (Masschelein et al. 1985)

Run Number	Percent Recycled	Color Ext. U.V. 250 nm (m^{-1})	Suspended Solids mg/L	Dissolved Al^{3+} mg/L	Final pH
1	0	3.78	6.1	0.071	7.41
	25	3.35	3.35	0.103	7.64
	50	5.0	5.0	0.088	7.78
2	0	5.3	3.72	0.141	7.52
	45-50	4.76	2.90	0.110	8.04
3	0	4.46	3.05	0.110	7.48
	50	5.25	5.3	0.247	8.1
	100[2]	7.38	12.1	2.280	8.4

[1]All values reported are averages of three tests
[2]Experience during algal bloom period (June 1983)

was compared to that obtained using 23 mg/L of commercial alum, 3 mg/L of activated carbon and 5 mg/L of chlorine. Water treated with recoveredcoagulant required pH adjustment with 0.15 g/L of sulfuric acid. A comparison of these finished waters is presented in Table 43. Aside from an increase in turbidity from 0.5 to 1.0 JTU, measured parameters were essentially equal.

Recovered Coagulant Trace Contaminants

Organic Matter and Color. In laboratory-scale testing of aluminum recovery using NaOH, Isaac and Vahidi (1961) concluded that organic matter bound with the sludge was much more soluble in the alkaline extraction procedure as compared to extraction using acid. Above pH 8, alkali soluble humus was thought to be dissolved. Although no data were presented, color associated with these organics was reportedly high in the recovered supernatant liquid.

TABLE 43

Finished Water Quality Following Jar-Tests Using Commercial and Recovered Coagulants (Wang and Yang 1961)

	Finished Water Quality	
Parameter	Commercial Alum	Recovered Sodium Aluminate
Turbidity, JTU	0.5	1.0
Color, CU	5.0	5.0
pH	7.1	6.4
Total Hardness, mg/L of $CaCO_3$	158	160
Coliform, number/100 mL	0	0
Aluminum, mg/L	<10	<10
Iron, mg/L	0.1	<0.1

Trace Contaminants in Alkali Recovered Contaminants. Masschelein et al. (1985) reported an extensive study into the metal contaminant concentrations in coagulants recovered using alkaline extraction. Metal concentrations as a function of pH resulting from treatment with NaOH are presented in Figure 34. In the previously described optimum pH range of 11.4 to 11.8 for NaOH, iron was the most prevalent contaminant found at a concentration of about 0.3 to 0.5 mmole/mole aluminum (621-1034 mg/kg Al). In order of highest to lowest concentration, nickel, cobalt, copper, manganese and chromium were also detected in the recovered coagulant in concentrations ranging from a high of 0.15 mmole/mole Al to a low of 0.02 mmole/mole Al.

Metal concentrations as a function of pH using CaO as an extractant are presented in Figure 35. In the optimum pH range of 11.2 to 11.6, iron was the most abundant contaminant at a concentration of about 0.085 mmole/mole (180 mg/kg Al). In order of decreasing concentration, nickel, cobalt, copper, chromium and manganese were found at concentrations from 0.035 mmole/mole to 0.002 mmole/mole aluminum. Masschelein et al. (1985) concluded that coagulant recovery using $Ca(OH)_2$ resulted in impurity concentrations 10 to 15 times lower than found using NaOH, making lime recovery more attractive where heavy metals were of concern.

These contaminant concentrations in the optimum pH ranges for recovery using NaOH and $Ca(OH)_2$ are presented in Table 44 normalized to aluminum concentration (i.e., mg/kg Al) for comparison to the Water Chemicals Codex (National Research Council 1982) standards presented earlier. Iron was the most prevalent compound in the extracts at concentrations of 620 Al to 1034 mg/kg Al in the NaOH extract and 176 mg/kg Al in the CaO-recovered coagulants. Manganese and chromium, which appear to be problematic in acid recovered coagulants, were detected at low concentrations. Manganese was reported at 81 mg/kg Al and 4 mg/kg Al, while chromium was detected at 38.5 mg/kg Al and 7.7 mg/kg of Al, respectively, for NaOH- and CaO-recovered coagulants. These concentrations are significantly lower than those reported in acidic-recovered coagulants. Further contaminants in CaO-recovered coagulants were nearly an order of magnitude lower than those detected in NaOH-recovered coagulants.

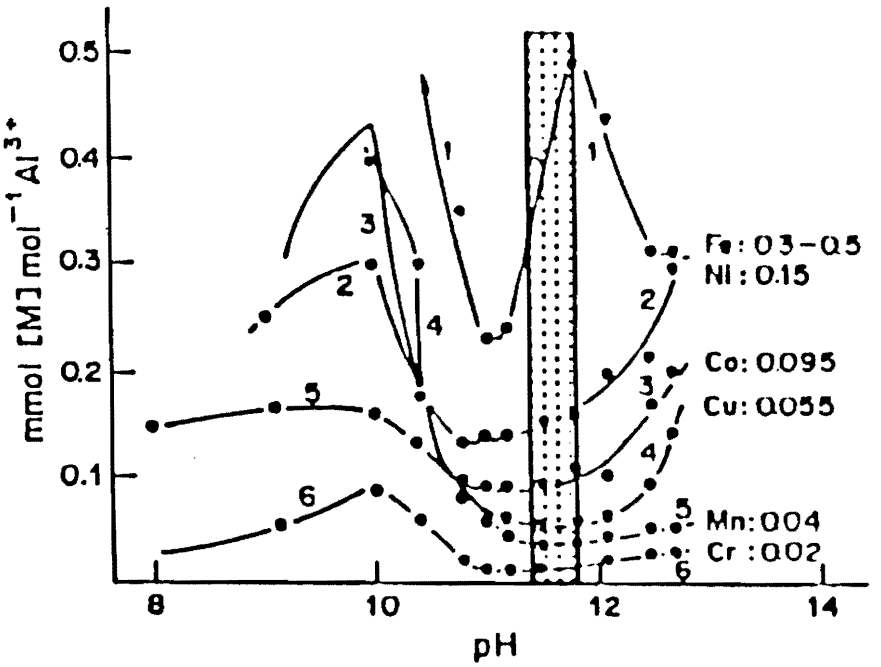

FIGURE 34. Trace Metals Contained in NaOH-Recovered Coagulants as a Function of Suspension pH (Masschelein et al. 1985)

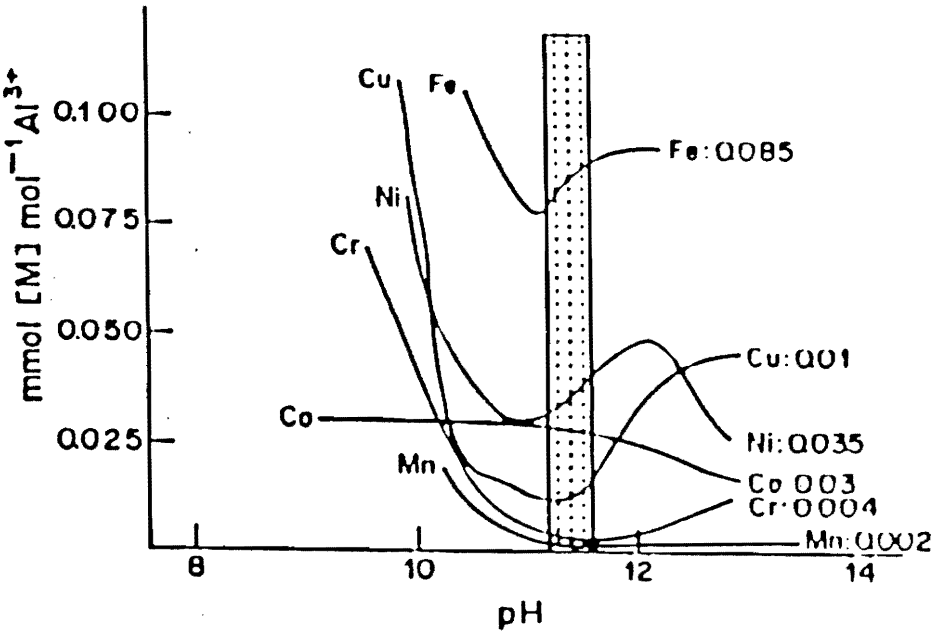

FIGURE 35. Trace Metals Contained in CaO-Recovered Coagulants as a Function of Suspension pH (Masschelein et al. 1985)

TABLE 44

Trace Contaminant Concentrations in Alkaline Recovered Aluminum Extracts at Reported Optimum pH Conditions (Masschelein et al. 1985)

Compound	Codex* mg/kg Al	NaOH Extraction mmole/mole	NaOH Extraction mg/kg Al	$Ca(OH)_2$ Extraction mmole/mole	$Ca(OH)_2$ Extraction mg/kg Al
Fe	**	0.3-0.5	620-1034	0.085	176
Ni	**	0.15	326	0.035	76
Co	**	0.095	207	0.03	65
Cu	**	0.055	129	0.01	23.5
Mn	**	0.04	81	0.002	4
Cr	330	0.02	38.5	0.004	7.7

*Water Chemical Codex (National Research Council 1982)
**No criteria provided for these metals

Sludge solids concentration had a profound impact on contaminants in recovered coagulants, as well as recovered coagulant concentrations. The percent of total aluminum in recovered supernatant as a function of initial suspended solids concentration was used to find an optimum solids concentration for coagulant recovery (Figure 32). Below about 12 g/L of suspended solids, aluminum recovery was at a maximum for extraction using both $Ca(OH)_2$ and NaOH. Percent of the total sludge metals found in the recovered supernatant were also examined in the same manner. Metal concentrations in the recovered coagulant as a function of initial sludge solids concentration (Figure 36) showed a dramatic increase in contaminant levels as the solids concentration fell below about 16 g/L. Considering both aluminum recovery and contaminants in the recovered coagulants lead the investigators to conclude that an optimum solids concentration of 15-20 g/L existed, where aluminum could be maximized while contaminants were minimized for this particular sludge.

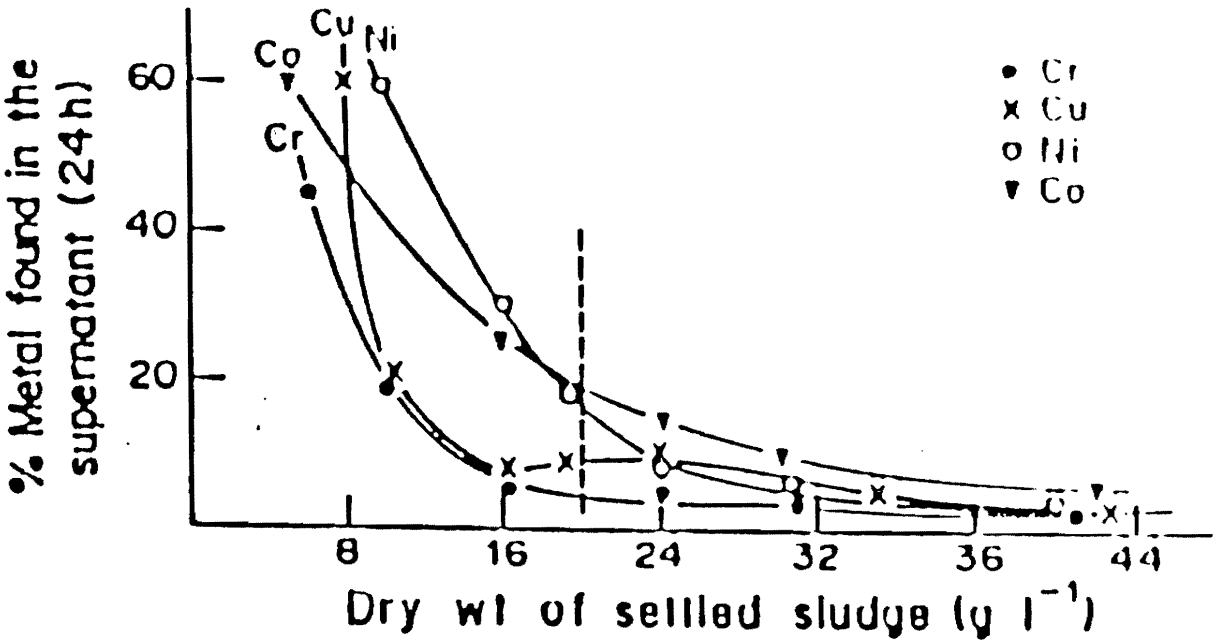

FIGURE 36. Percentage of Total Sludge Metals Contained in NaOH—Recovered Coagulants as a Function of Initial Sludge Suspended Solids Concentration (Masschelein et al. 1985)

Thickening and Dewatering of Residual Sludge Solids

Masschelein et al. (1985) examined the volume of recoverable supernatant as a function of initial suspended solids concentration, as presented earlier in Figure 28. Following 24 hours of settling, recoverable supernatant liquid ranged from about 10 percent of the total volume at a solids concentration of 42 g/L to about 90 percent at a solids concentration of 6 g/L. Results were almost identical for both NaOH- and $Ca(OH)_2$-treated sludges. The results presented are those obtained at the previously specified optimum pH for each recovery process (i.e., 11.4 for CaO and 11.6 for NaOH). At the optimum solids concentration for coagulant recovery, 60 percent volume reduction was experienced using CaO, while 50 percent volume reduction was experienced using NaOH. Reductions in residual sludge solids required to be dewatered and disposed of was not reported.

Slechta and Culp (1967) reported 90 percent recovery at pH 12 using NaOH. The 90 percent recovery was based on supernatant liquid recovery assuming complete aluminum dissolution (i.e., 90 percent volume reduction of the sludge). As presented earlier in Figures 30 and 31 for NaOH and $Ca(OH)_2$, respectively, filtration techniques for supernatant recovery proved more efficient than did gravity sedimentation. A maximum of 90 and 35 percent supernatant recovery was possible using NaOH and $Ca(OH)_2$, respectively, while only 75 and 25 percent supernatant recovery was possible using gravity sedimentation.

IRON RECOVERY FROM WATER TREATMENT PLANT SLUDGES

Use of iron salts in the treatment of drinking water has been limited due primarily to a previous limit of 0.3 mg/L placed on iron in finished waters by the USPHS Drinking Water Standards (USPHS 1965) and the potential for "red water" problems caused by iron precipitation in distribution systems. Due to the limited use of iron coagulants in the water treatment industry and the lack of research into iron recovery, there are few data available on the recovery of iron from iron hydroxide sludges. However, current concern over the health impacts of residual aluminum may result in iron salts being considered further for use as primary coagulants, increasing the potential for recovery of the iron coagulants.

OPERATIONAL VARIABLES AFFECTING IRON RECOVERY

A number of parameters have been identified as having a significant impact of the feasibility of iron recovery. These include: stoichiometry of iron dissolution; type of extractant; pH of acidified suspensions; and reaction kinetics. Acids and reducing agents have been applied in experimental investigations for iron recovery, while alkaline extraction of iron has not been reported in the literature.

Stoichiometry of Acidic Iron Recovery

Iron is a hydrolysing metal with a chemistry similar to that of aluminum, making much of the previous information on alum sludges pertinent to iron sludges. Sulfuric acid and hydrochloric acid have been investigated for use in the acidic extraction of iron from iron hydroxide sludges.The stoichiometry of iron coagulation and sulfuric acid recovery of ferric hydroxide with sulfuric acid can be described, respectively, in equations 7 and 8:

$$Fe_2(SO_4)_3 \cdot 3H_2O + 3Ca(OH)_2 \Rightarrow 3CaSO_4 + 2Fe(OH)_3 \cdot 3H_2O \quad (7)$$

$$2Fe(OH)_3 \cdot 3H_2O + 3H_2SO_4 \Rightarrow Fe_2(SO_4)_3 + 9H_2O \quad (8)$$

The stoichiometric acid dose is 1.5 mole H_2SO_4/mole of Fe, equivalent to an H_2SO_4 requirement of 2.63 kg/kg Fe or 0.91 kg/kg $Fe(OH)_3 \cdot 3H_2O$. The reaction between iron hydroxide and hydrochloric acid may be written as follows.

$$Fe(OH)_3 \cdot 3H_2O + 3HCl \Rightarrow FeCl_3 + 6H_2O \quad (9)$$

From this equation, the stoichiometric hydrochloric acid dose would be 3 mole/mole Fe, which is equivalent to 1.96 kg/kg Fe or 0.68 kg/kg $Fe(OH)_3$ $3H_2O$.

Acid Type: H_2SO_4 versus HCl

King et al. (1975) presented data relating iron recovery to pH using both sulfuric and hydrochloric acids. An iron sludge obtained from a water treatment plant using ferric sulfate as a primary coagulant was tested. As presented in Figure 37, slightly more iron was recovered using sulfuric acid above pH 2, while below this, both proved to be about equal. Recovery using sulfuric acid was found to be more feasible based on economic considerations, as expressed previously in the alum recovery process.

In studying removal of phosphates and metals from anaerobically digested sludge conditioned with ferric chloride and lime, Scott and Horlings (1975) conducted kinetic tests on iron extraction using varying amounts of sulfuric and hydrochloric acid, as presented in Figure 38. The percent excess expressed was based on stoichiometric acid requirements of calcium, iron, aluminum and zinc ions in the sludge. In all extraction tests, pH was 1.5 or less. It was concluded that H_2SO_4 and HCL performed equally well. H_2SO_4 was subsequently used for further study based on lower costs, removal of $CaSO_4$ and improved filterability of the residual sludge. Scott et al. (1980) reported that Oliver and Carey (1976) extended the work of Scott and Horlings (1975) and concluded that there was little difference in extraction performance of nitric, hydrochloric, and sulfuric acids.

Evaluation of Experimental Sulfuric Acid Additions

In a bench scale study, White and White (1984) reported on one attempt at iron

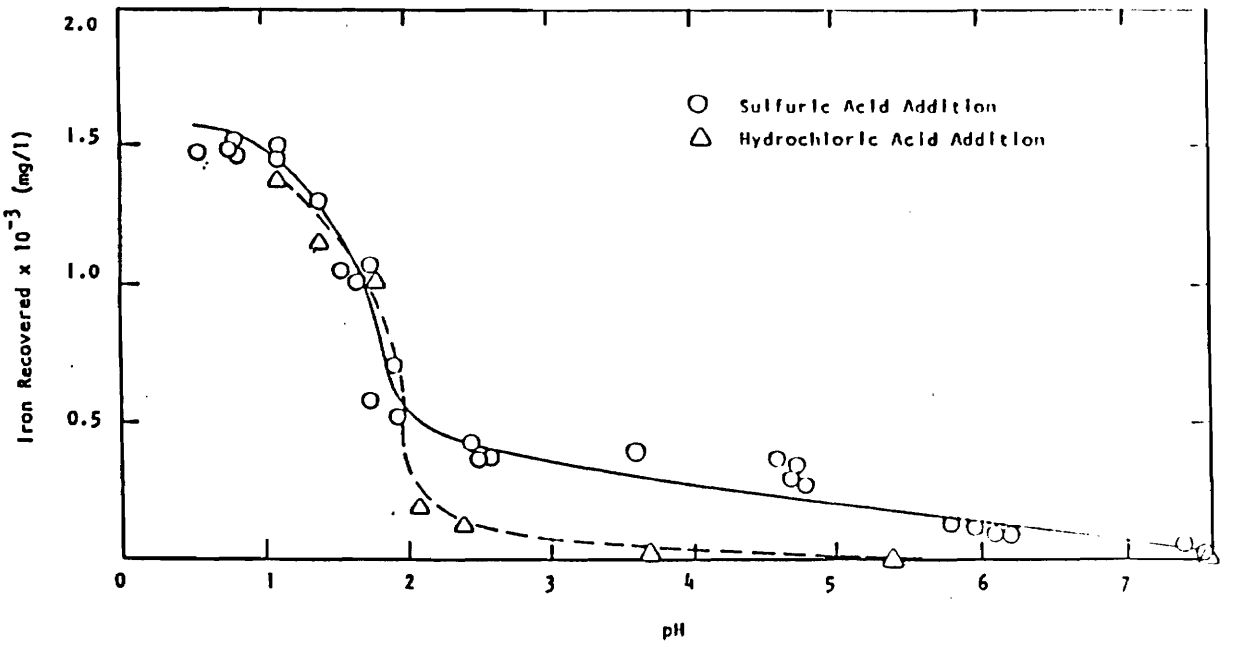

FIGURE 37.. Recovered Iron Concentrations as a Function of Suspension pH Following Addition of Sulfuric and Hydrochloric Acid (King et al. 1975)

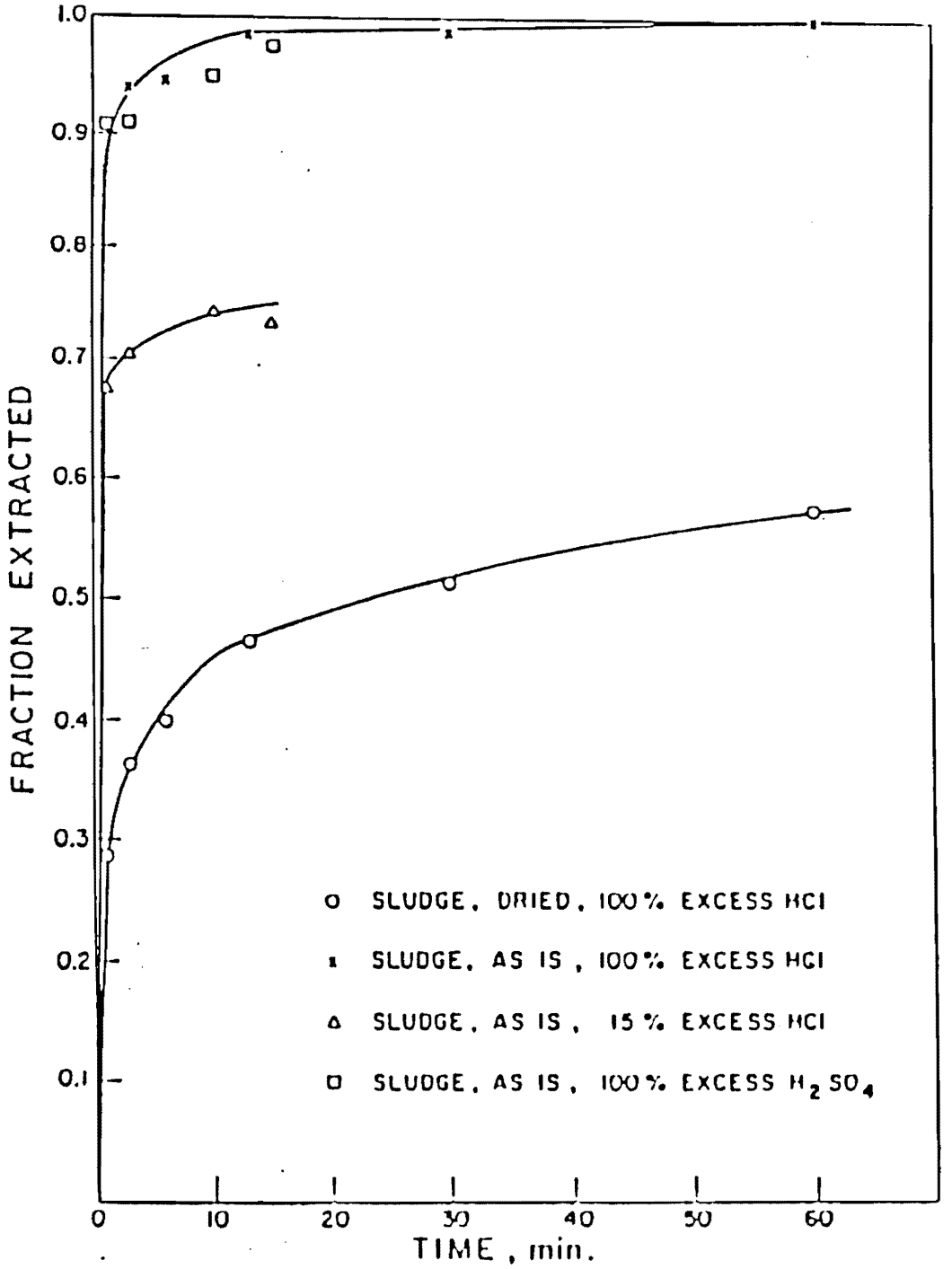

FIGURE 38. Fraction of Total Iron Extracted from Iron-Conditioned Anaerobically-Digested Sewage Sludge. Percent Excess HCl and H_2SO_4 Calculated for Dissolution of Iron as $Fe(OH)_3 \cdot 3H_2O$ to Fe^{3+}, e.g., 100% excess equals twice stoichiometric acid addition (Scott and Horlings 1975)

recovery. An iron sludge containing 4.5 percent solids was treated with 6.2-mL H_2SO_4/L of sludge (approximately 0.25 g H_2SO_4/g solids) to attain a pH of 2.5. No report was made as to percent recovery or concentration of the recovered coagulant. King et al. (1975) reported iron recovery from a ferric sludge as a function of pH (Figure 37). The sludge examined had a total solids concentration of 40.8 g/L with 2.35 g/L as iron. Use of sulfuric acid resulted in a recovered solution containing iron at a concentration of 0.5 g/L at a pH value of 2.0, equivalent to about 21 percent recovery of aluminum. As the recovery pH decreased to a value of about 1, soluble iron concentrations increased to about 1.5 g/L or 64 percent recovery.

Evaluation of Experimental Hydrochloric Acid Additions

Pigeon (1976) reported iron recovery from a synthetic, iron-hydroxide sludge, as well as two water-treatment-plant sludges obtained from the St. Louis County Water Company and the Kingsport, TN water treatment plants. Ferric chloride was used to generate the synthetic sludge, while both water treatment plants used ferric sulfate as primary coagulant. Characteristics of the sludges were presented earlier in Table 9. Both water treatment sludges were diluted to 2.0 percent solids prior to analysis. Volatile solids were 14 percent and 5 percent of the total solids for the Kingsport and St. Louis county sludges, respectively, while the suspended solids were 99.5 percent and 99.0 percent of the total solids, respectively. The iron content of the Kingsport sludge was 4120 mg/L as Fe at a pH value of 7.4, while the St. Louis county sludge contained 930 mg/L as Fe and had a pH value of 8.5. Specific resistance and settling velocity were not reported.

Hydrochloric acid was used to acidify the sludge samples to pH values ranging from 1.0 to 5.0. The amount of hydrochloric acid required to obtain a particular pH value is presented in Figure 39 for the Kingsport and St. Louis County sludge. The St. Louis County sludge exhibited a much higher acid requirement as compared to the other two sludges. For example, to attain a pH of 2.0, about 85 meq/L of HCl were required for the Kingsport sludge, while greater than 300 meq/L were required for the St. Louis County sludge. It was concluded that the increase demand was caused by the presence of carbonates or

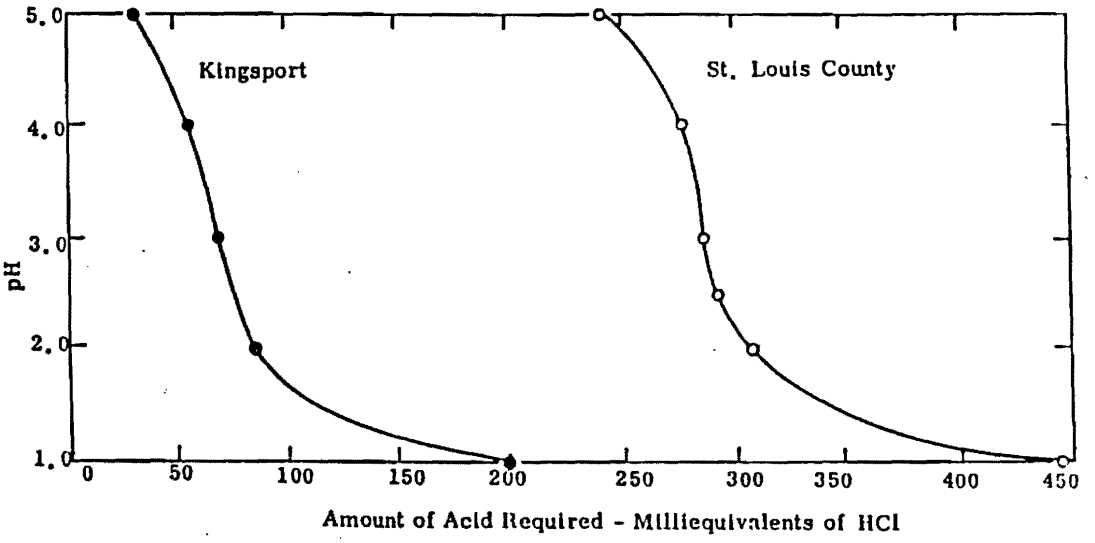

FIGURE 39. pH of Acidified Sludge Suspensions as a Function of Hydrochloric Acid Addition for Ferric Hydroxide Sludge Suspensions (Pigeon 1976)

lime in the St. Louis County sludge, as evidenced by vigorous bubbling upon acidification.

The percentage of the stoichiometric amount of hydrochloric acid added for iron recovery is presented in Figure 40 as a function of the percentage of total iron recovered for the Kingsport and synthetic sludges. Data collected by Weeks (1974) while testing iron recovery from the Kingsport sludge is also presented in this figure. Although Weeks (1974) was able to recover greater than 60 percent of the total iron, a plateau was reached where further recovery was not possible with addition of more acid, much the same as experienced by Pigeon (1976). Total iron recovered as a function of pH is presented in Figure 41, while the percentage of iron recovered is presented in Figure 42 as a function of pH. Iron concentrations were determined after 10 min of mixing following acid addition.

The Kingsport sludge, with a total iron content of 4120 mg/L of Fe, yielded a maximum recovered iron concentration of about 900 mg/L of Fe (26 percent recovered) below a pH value of about 3. Recovery from the St Louis County sludge (930 mg/L total Fe) consistently increased as the pH dropped to 1.0, where 450 mg/L of Fe (55 percent) was recovered. Iron recovery from the synthetic sludge exceeded that of the water treatment plant sludges below a pH of about 3.0, with 95 percent recovery at a pH value of 1.0.

Although a 10-min detention time was used in the above tests, subsequent kinetic tests showed that maximum dissolution of iron occurred about 30 min following acid addition, as presented in Figure 43. During the kinetic test, a recovered iron concentration of about 950 mg/L of Fe was achieved with the Kingsport sludge after 10 min of mixing, increasing by 26 percent to about 1200 mg/L of Fe following 30 min of mixing. A significant increase in recovered iron concentration could therefore be expected in the earlier tests if the mixing time had been 30 min.

Sulfide reduction of the sludges was also tested to determine its impact on recovery. Sludge samples were acidified to a target pH value with HCl prior to addition of a stoichiometric amount of Na_2S. Figure 44 presents data

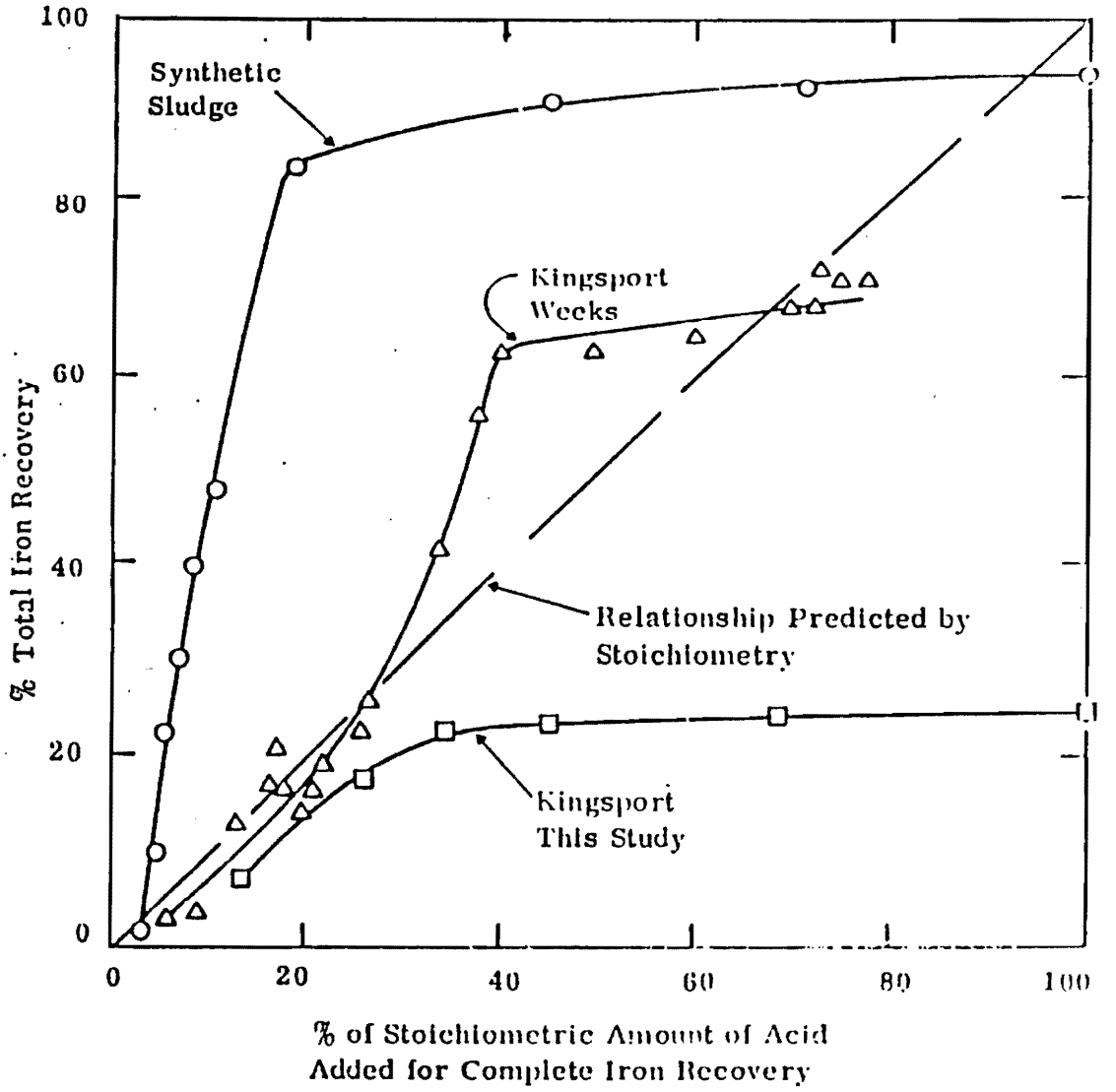

FIGURE 40. Percentage of Total Iron Recovered as a Function of the Percent of Stoichiometric Acid Addition Based on Dissolution of $Al(OH)_3 \cdot 3H_2O$ to Al for Synthetic and Kingsport Sludges (Pigeon 1976)

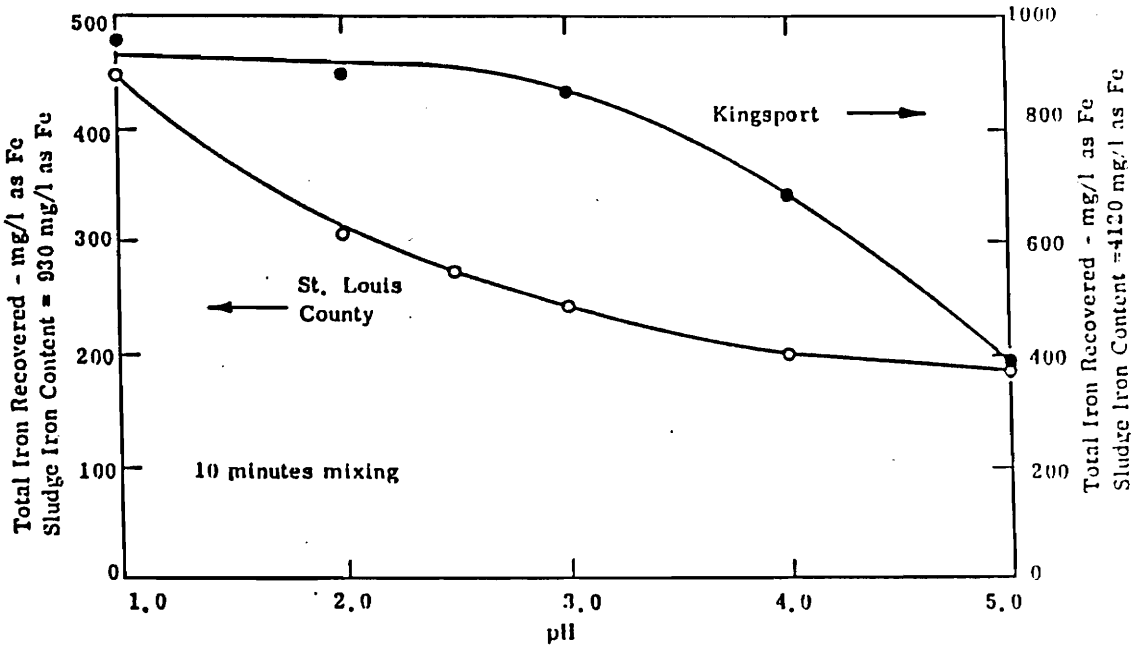

FIGURE 41. Recovered Iron Concentration as a Function of Suspension pH following HCl Addition to St. Louis County Sludge (Total Fe = 4120 mg/L as Fe) and Kingsport Sludge (Total Fe = 930 mg/L as Fe) (Pigeon 1976)

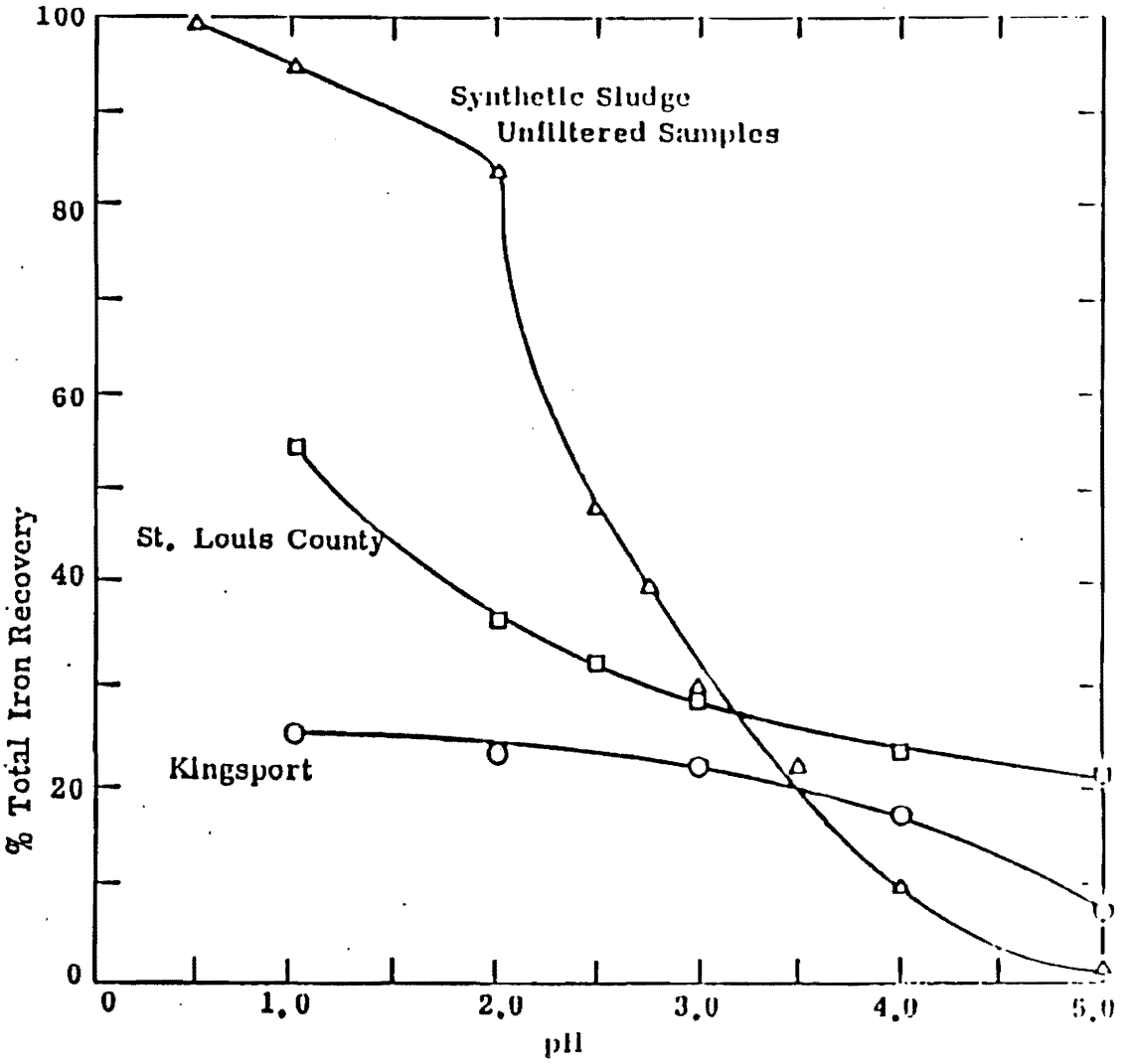

FIGURE 42. Percentage of Total Sludge Iron Recovered as a Function of Suspension pH (Pigeon 1976)

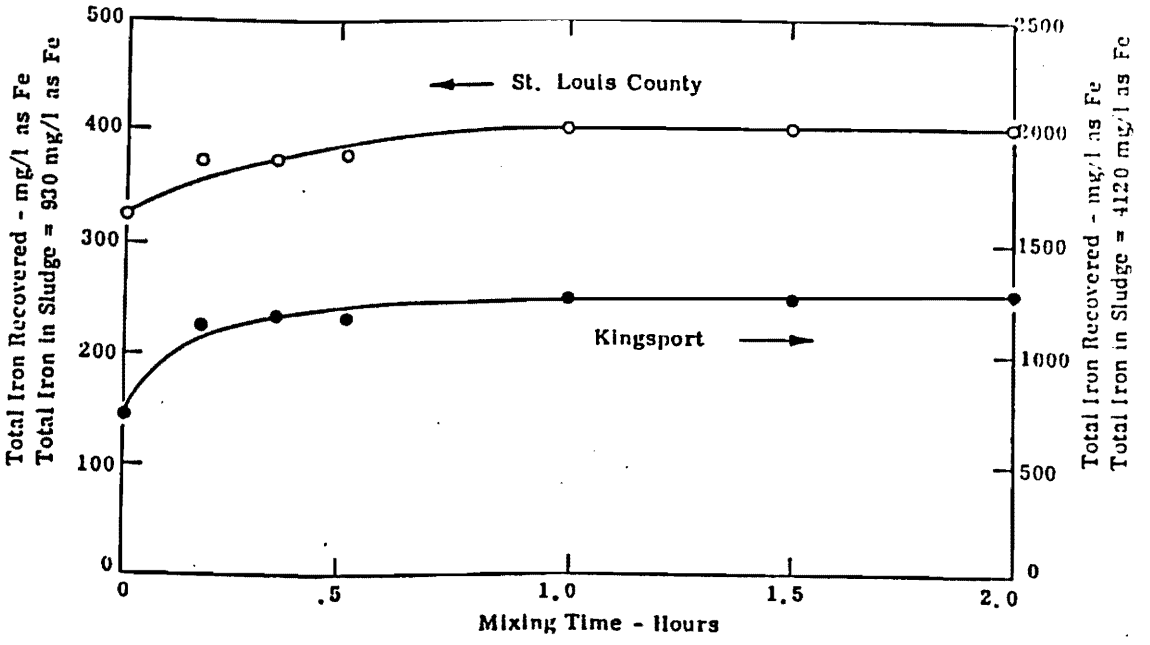

FIGURE 43. Kinetic Tests for Iron Recovery Following Hydrochloric Acid Addition (Pigeon 1976)

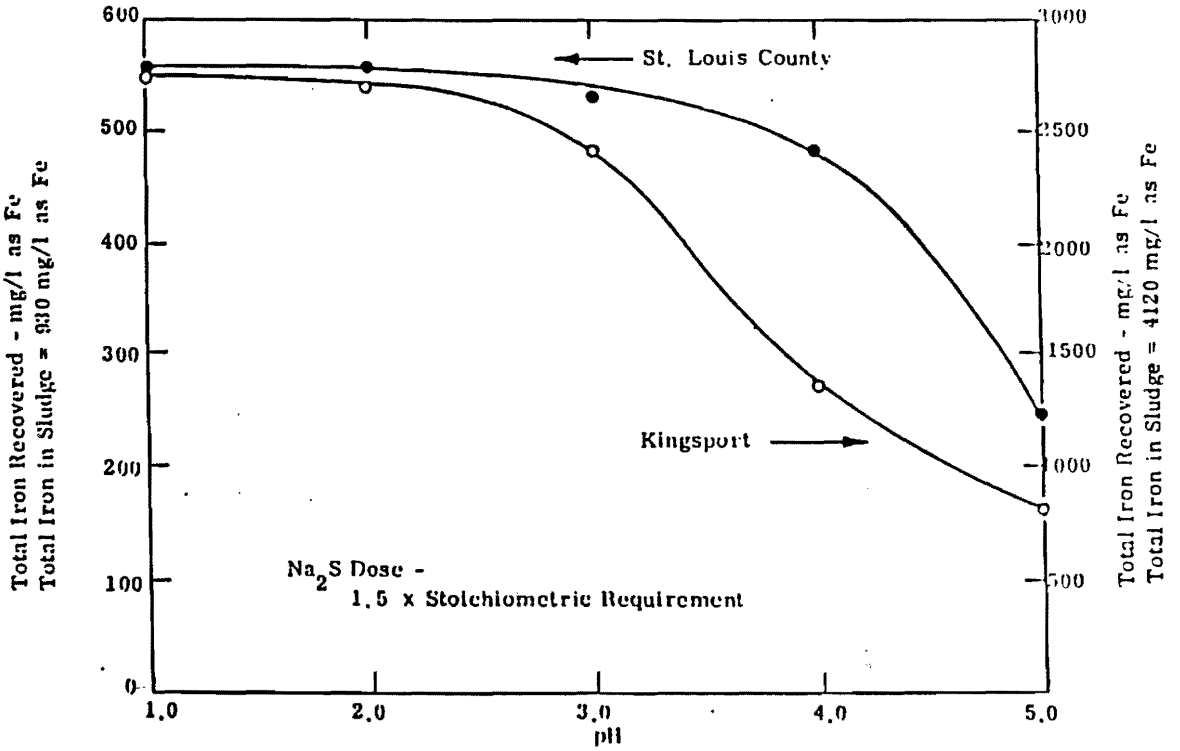

FIGURE 44. Results of Sulfide Reduction on Iron Dissolution as a Function of Suspension pH Following Hydrochloric Acid Addition (Pigeon 1976)

obtained using a sodium sulfide dose of 1.5 times the stoichiometric requirement based on the total iron concentration. Maximum iron dissolution for both sludges occured at a pH of about 3.0, below which little more iron could be recovered. Maximum recovered iron concentrations were 560 mg/L of Fe and 2750 mg/L of Fe at pH 3 for the St. Louis and Kingsport sludges, respectively. This corresponds to an increase in recovered iron concentrations following reduction with Na_2S of 24 and 200 percent, respectively, over that obtained using acid alone. Use of sulfide can cause problems with H_2S formation, therefore reducing its potential as an extractant. Furthermore, reduction of iron to the more soluble ferrous state may impact the effectiveness of the coagulant for reuse as a coagulant in water treatment.

UTILITY OF RECOVERED IRON COAGULANTS

Settled water turbidity was used as a measure of recovered coagulant efficiency by Pigeon (1976) and compared to a standard coagulation curve developed using reagent-grade ferric chloride as coagulant (Figure 45). Coagulation tests were performed using a jar test apparatus with an autoburret to control the pH after coagulant addition. Iron solutions were recovered using HCl to adjust the pH to a value of 3, followed by Na_2S addition at 1.5 times the calculated stoichiometric dose. Coagulation tests using iron recovered from the St. Louis County sludge had a critical coagulation concentration (CCC) of about 13 mg/L, substantially lower than the 18 mg/L obtained using reagent-grade coagulant. Iron recovered from the Kingsport sludge was less effective with a CCC of about 26 mg/L. However, water treated with either recovered coagulant had a settled water turbidity equal to that of the reagent-grade coagulant.

In studying iron recovery from a water treatment plant using ferric sulfate as primary coagulant, King et al. (1975) compared commercial and recovered coagulants on kaolinite and natural water suspensions. Although the characteristics of the raw water varied from that of the kaolinite suspension, similar results were obtained for each, as shown in Figures 46 and 47. Although the critical coagulation concentration was lower with the recovered

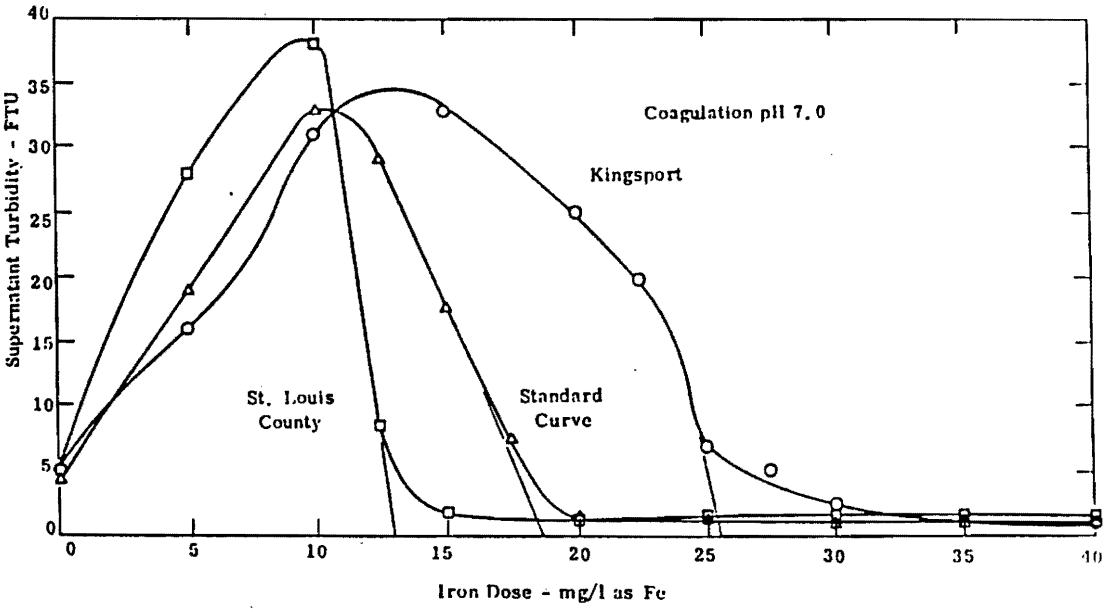

FIGURE 45. Residual Turbidity as a Function of Recovered and Commercial Iron Doses (Pigeon 1976)

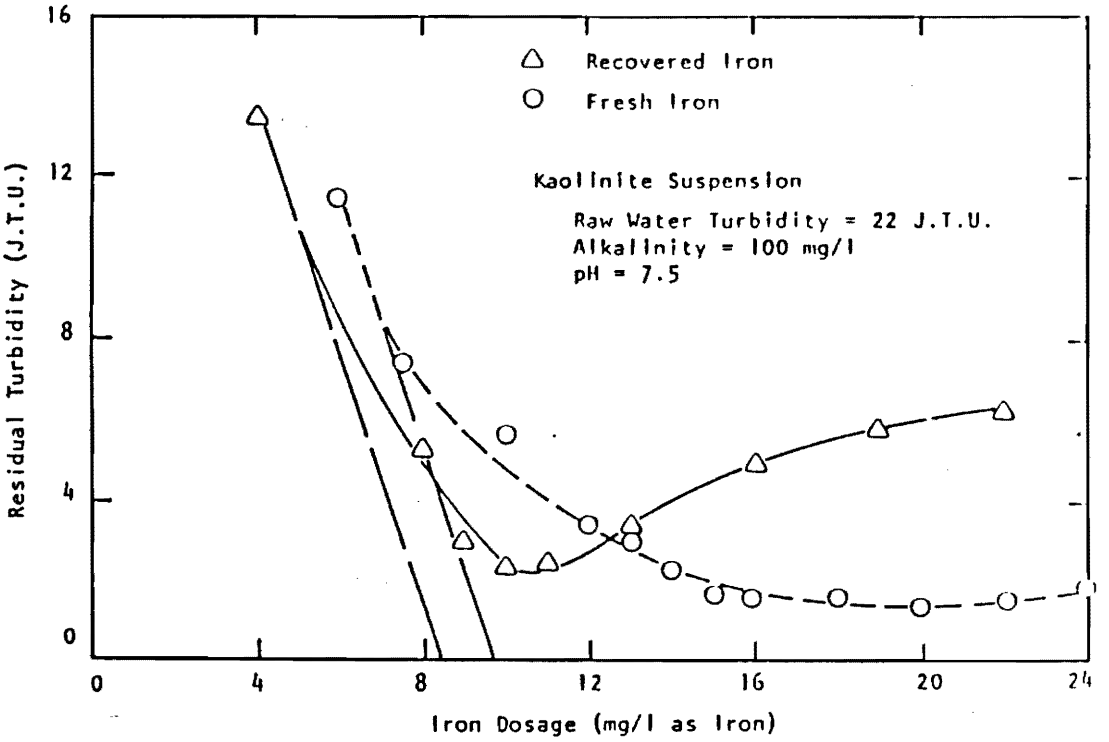

FIGURE 46. Residual Turbidity and Critical Coagulation Concentration Following Coagulation of an Artificial Kaolinite Suspension Using Commercial and Recovered Coagulants (King et al. 1975)

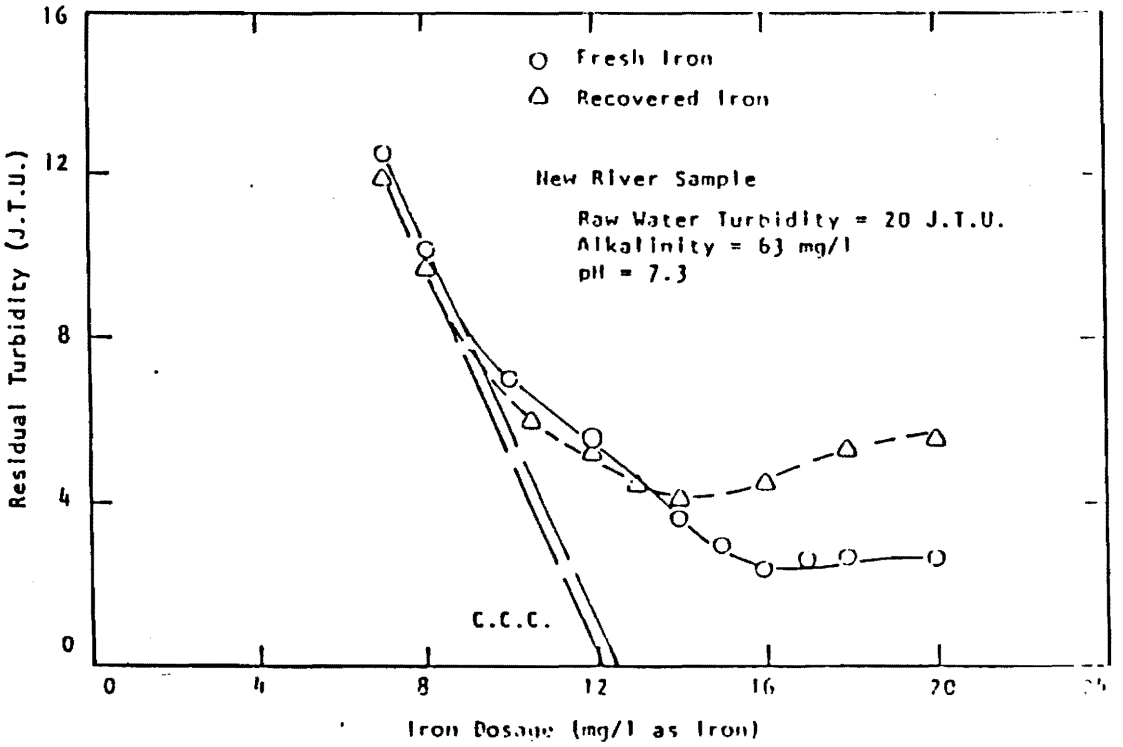

FIGURE 47. Residual Turbidity and Critical Coagulation Concentration Following Coagulation of a Natural River Water Using Commercial and Recovered Coagulants (King et al. 1975)

coagulant on both the kaolinite and raw water sources, commercial iron had a larger effective dose range for turbidity removal in coagulation tests using the kaolinite suspension. Coagulation of the raw water showed much the same trend, although the recovered iron solution was never able to effectively remove turbidity as compared to the reagent-grade iron solution. It should be noted that turbidity and pH of the suspensions compared above differed somewhat, with both being lower in the natural water suspension.

THICKENING AND DEWATERABILITY OF RESIDUAL SLUDGE SOLIDS

King et al. (1975) presented extensive data on dewaterability and thickening of acidified iron sludges. Thickening properties were characterized through the use of initial settling velocity, as presented in Figure 48. The initial settling velocity increased consistently with a decrease in pH, indicating an improvement in the settling characteristics of the sludge with increasing acid addition. Although no data were presented, King et al. (1975) also reported a dramatic reduction in the volume of residual solids to be dewatered after acidification. Specific resistance was used by King et al. (1975) as a measure of sludge filterability following acidification. As presented in Figure 49, dramatic improvements in filterability were demonstrated with a decrease in pH to approximately 2.0. Below a pH value of 2, filterability deteriorated considerably.

In treating an iron sludge with 4.5 percent dry solids, White and White (1984) reported an increase from 22 percent dry cake solids for non-acidulated sludge to 23 percent for acidulated sludge at pH 2.5. No data were presented for sludge volume reduction or reduction of total solids for disposal.

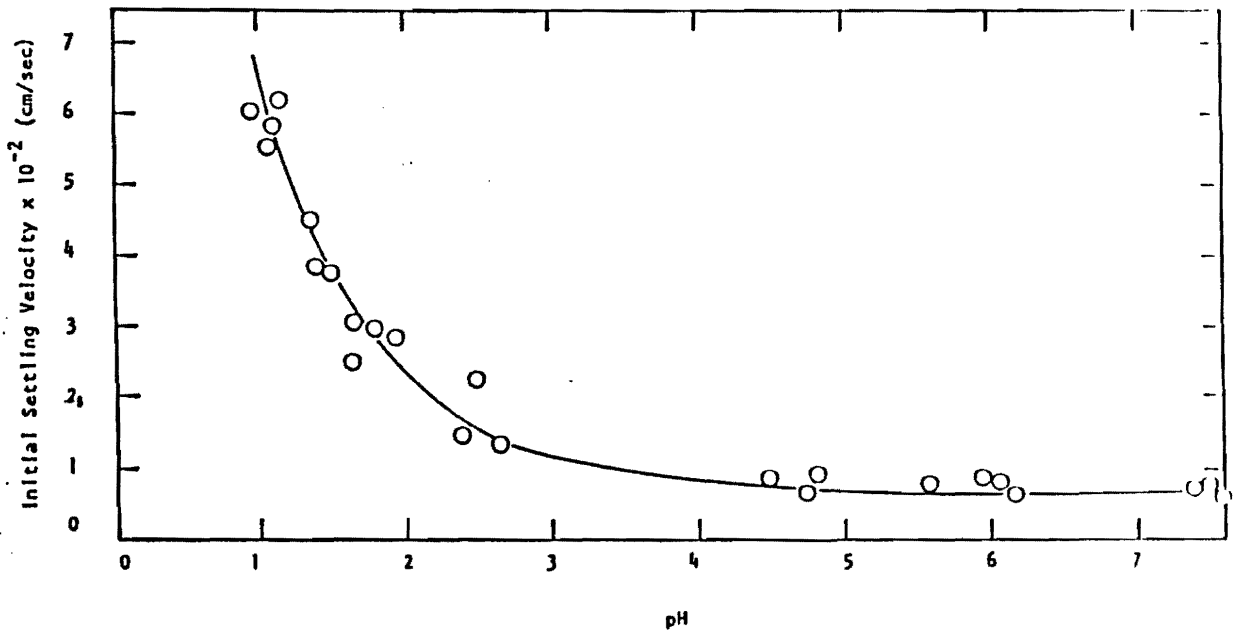

FIGURE 48. Initial Settling Velocity Following Acidification as a Function of Suspension pH (King et al. 1975)

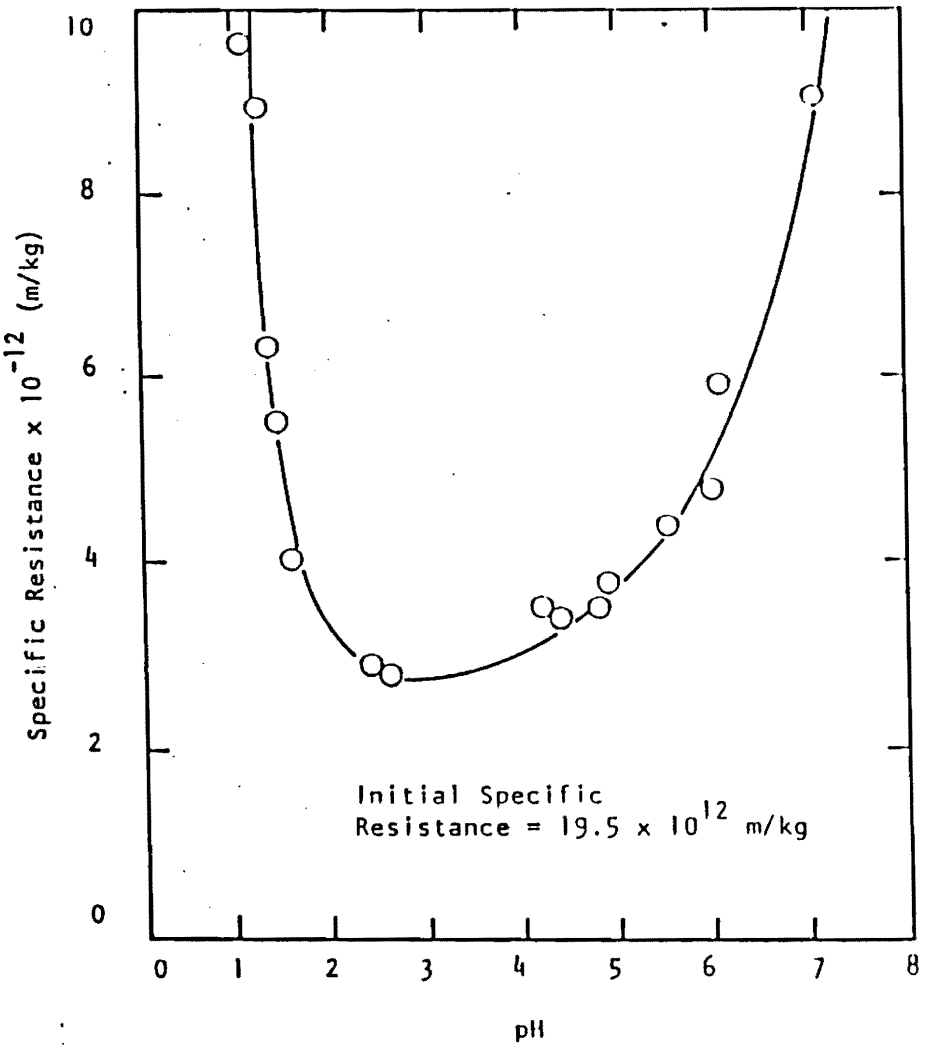

FIGURE 49. Specific Resistance Following Acid Addition as a Function of Suspension pH (King et al. 1975)

FULL-SCALE COAGULANT RECOVERY OPERATIONS

Although numerous laboratory- and pilot-scale investigations of coagulant recovery have been reported in the United States, full-scale evaluation and operation has been limited. Full-scale investigations of coagulant recovery and pilot-scale investigations utilized for full-scale design considerations are reported in Table 45. Roberts and Roddy (1960) and Bishop et al. (1987) have reported full-scale testing of coagulant recovery at Tampa, FL and Durham, NC, respectively. The results of these studies were presented earlier. Atlanta, GA conducted pilot-scale studies into coagulant recovery for potential incorporation into the North Area Water Treatment Plant. The results of this research are presented by Saunders (1989) and were discussed in detail earlier in this text. The objective of the remainder of this text is to present pilot- and full-scale design and operation investigations of coagulant recovery systems at Jersey City, NJ, in numerous plants in Japan and at Athens, TN and to present an operational and economic analysis of associated construction, operation and maintenance of coagulant recovery systems.

PILOT- AND FULL-SCALE INVESTIGATIONS OF COAGULANT RECOVERY

Fulton (1974) reported the design and construction of a coagulant recovery system at the Jersey City water filtration plant in New Jersey. A summary of the results of this research and a review of the subsequent United States Patent for coagulant recovery will be discussed, along with the coagulant recovery process operations at the Jersey City facility.

Japan has been utilizing coagulant recovery technology more than two decades for minimization of sludge volume and mass for disposal and recovery of the primary coagulant used in water treatment. Historical data from nine water treatment plants in Japan utilizing coagulant recovery were evaluated to provide insight into systems operation and performance based on many years of full-scale experience.

Although recovery of iron coagulants has received little attention in the past, Athens, TN has constructed and is currently operating a full-scale

TABLE 45

Pilot- and Full-Scale Investigations of Coagulant Recovery

Location	Coagulant	Comments	Reference
Tampa, FL	Aluminum	Pilot- and full-scale recovery with H_2SO_4	Roberts and Roddy 1960
Durham, NC	Aluminum	Full-scale evaluation of use of acidic coagulant recovery and dewatering with sand-drying beds	Bishop et al. 1987
Atlanta, GA	Aluminum	Pilot-scale evaluation of product quality of acidic extractant and pressure-filter dewatering of acidic and lime-treated sludge	Saunders 1989
Jersey City, NJ	Aluminum	Full-scale plant using Fulton process for acidic aluminum recovery	Fulton 1974
Japan	Aluminum	Fifteen plants constructed using acidic aluminum recovery between 1965 and 1972	Committee Report 1978
Athens, TN	Iron	Full-scale evaluation of acidic recovery and sludge dewatering	White, A.R. and White, P.M.R. 1984
Spartanburg, SC	Aluminum	Pilot-scale evaluation for economic evaluation of full-scale implementation	Jordan, Jones and Goulding, Inc. 1988

coagulant recovery facility for recovery of the primary coagulant, ferric sulfate, and minimization of sludge solids. Since initial construction and operation in 1985, the system has undergone numerous physical and operational changes for optimization of the coagulant recovery process. In 1988, the coagulant recovery process was used to treat all but one volume of waste sludge solids and the resulting recovered coagulants were utilized to treat about 35 percent of the potable water produced in 1988. Through the cooperation of plant personnel, operational and performance data (Camp 1989) were obtained and evaluated for both the coagulant recovery system and water treatment operations and performance. Although capital costs of the system were not available, a detailed analysis of total chemical costs for coagulant recovery and water treatment using commercial and recovered coagulants was conducted. The results of this investigation are presented in detail.

The American Water Works Service Company (1987) investigated numerous water treatment sludge conditioning and dewatering systems and evaluated each with respect to operational and economical considerations. Required land area, total sludge production, capital costs, operating and maintenance costs and a 20-year present worth analysis are presented for a comparison of the alternatives investigated. Spartanburg, SC has recently conducted an investigation into full-scale implementation of a coagulant recovery system at the R. B. Simms Filtration Plant. A pilot-study of solids handling incorporating coagulant recovery was conducted by Jordan, Jones and Goulding, Inc. (1988). The results were used to conduct a complete economic analysis of construction, operation and maintenance costs. A summary of the results of this research and final economic analysis of the proposed full-scale coagulant recovery system are be presented.

JERSEY CITY COAGULANT RECOVERY SYSTEM

Fulton (1974) conducted research into the development of an acidic aluminum-recovery process in conjunction with Metcalf and Eddy, Inc. The culmination of this work was a U.S. patent (No. 3,959,133) of the alum recovery and waste disposal system presented in Figure 50 and the design and construction of a coagulant recovery system at the Jersey City, NJ water treatment plant. The patented system consisted of a three-phase operation of a filter press

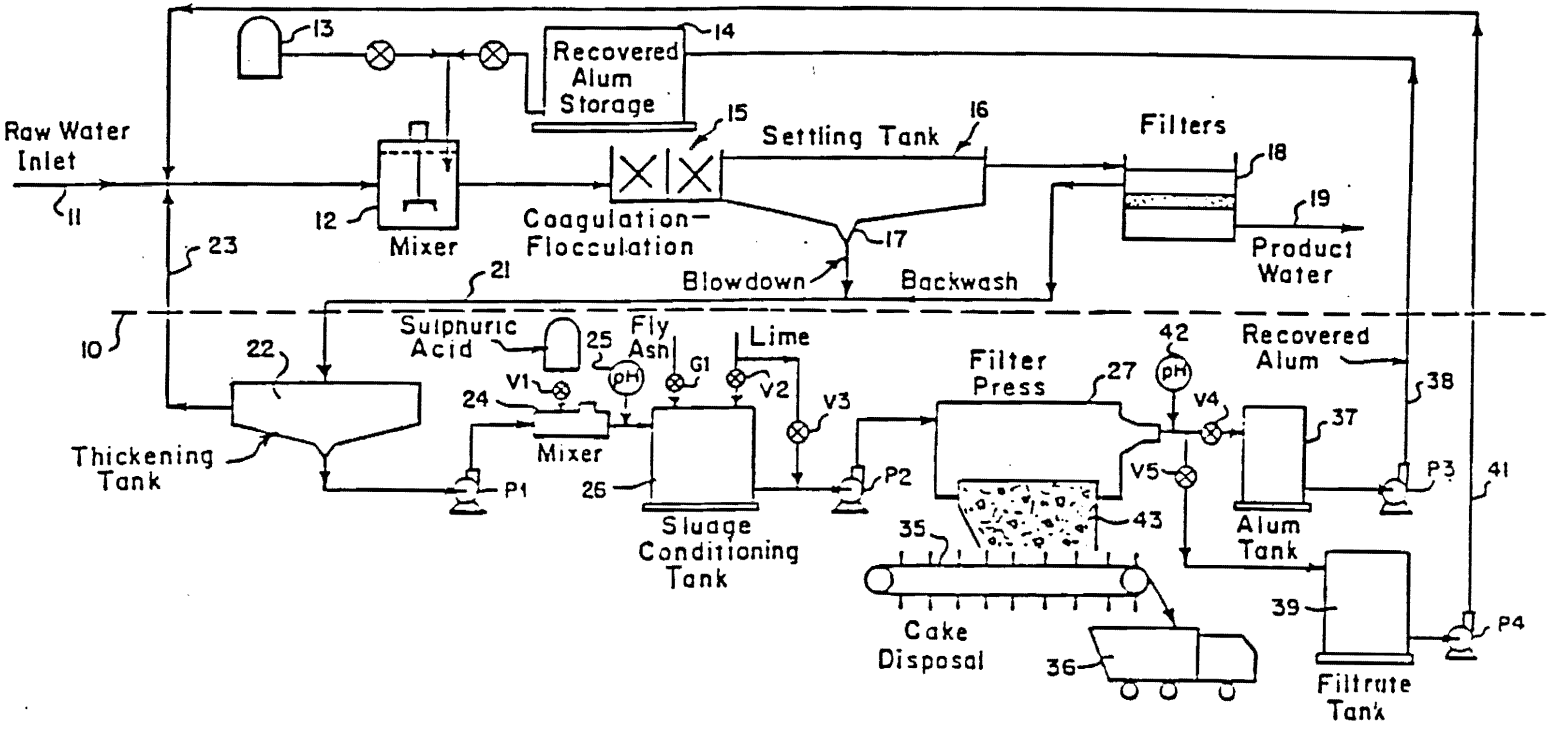

FIGURE 50. Schematic Diagram of Coagulant Recovery System Developed by Fulton (1974) and Contained in United States Patent No. 3,959,133

dewatering system. During the alum recovery phase, alum sludge solids produced in the water treatment system are thickened and conditioned with concentrated sulfuric acid. The acidified sludge solids are fed directly to a filter press to dewater the acidic sludge solids, while the filtrate from the filter press is recovered as a coagulant for reuse in the water treatment system. At the end of a filter press cycle, the supply of acid-conditioned sludge solids is curtailed and a lime slurry is passed through the filter press to neutralize acidified sludge solids contained in the chambers. Periodically, contaminants may build up in the recovered coagulant requiring use of the sludge dewatering system without acid conditioning of sludge solids to enable wastage of the sludge solids produced from the use of recovered coagulants. After the recovered coagulant volume has been used, commercial alum is employed for a time adequate for dewatering of the sludge solids produced from recovered coagulant use, i.e., wastage of all sludge solids produced from use of recovered coagulants and the associated build-up of contaminants from the recovered coagulants. The system can then be converted to the alum recovery operational mode.

The results of the research and details of the proposed recovery process by Fulton (1974) resulted in the design of a coagulant recovery system for the 303,000 m^3/d (80 mgd) Jersey City, NJ water filtration plant. Fulton (1974) concluded that a minimum sludge solids concentration of about 2 percent should be maintained and the sludge solids should be conditioned to a pH value of 2.0 for adequate operation of the coagulant recovery system. The sludge processing facilities were identical to a standard filter press dewatering system having a sludge conditioning tank for sulfuric acid contact and the capability of lime and diatomaceous earth conditioning of sludge solids. One of the three filter presses provided was constructed of stainless steel with plastic-covered cast iron press plates to minimize acid corrosion problems. The sludge conditioning tank was also constructed of acid-resistant materials with stainless steel pipe connections to the filter press. The system utilized in-line addition of concentrated sulfuric acid upstream of an in-line mixer at the inlet to the sludge conditioning tank.

The coagulant recovery system was fully designed and constructed at the Jersey City plant and all components of the system, with the exception of sulfuric

acid storage and addition, were tested and reported to be operational. However, the coagulant recovery facility was never put into operation due to problems encountered with the acid storage system. Publications in the literature have referenced the Jersey City facility as one location of full-scale implementation, but no operational data were reported or published. Lack of such information has lead many to conclude that the coagulant recovery system was not successfully implemented. However, the coagulant recovery system at the Jersey City facility was apparently not certified by the design engineers for operation due to problems encountered onsite during initial startup, and the system was never put into operation or tested at the full-scale level.

FULL-SCALE COAGULANT RECOVERY EXPERIENCE IN JAPAN

Increasing regulations governing disposal of water treatment plant sludges in Japan in the late 1960's and early 1970's resulted in the design and construction of many full-scale coagulant recovery systems. Coagulant recovery was employed for sludge conditioning prior to dewatering and ultimate disposal, resulting in the reduction of sludge volume and mass for disposal and the recovery of a coagulant for reuse at the water treatment facility (AWWA Committee Report 1978; Westerhoff and Daly 1974). To investigate the use of coagulant recovery in Japan, extensive operational and process information was obtained from The Department of Sanitary Engineering at The Institute of Public Health in Japan for nine water treatment plants employing coagulant recovery. Data received indicates that some coagulant recovery facilities were in operation as late as 1987, but information regarding the current status of these systems is not available. However, one of the sites investigated in this report, the Senzou facility at Itami-City, has reportedly abandoned the process.

Through contact with Dr. Yasumoto Magara, Director of the Department of Sanitary Engineering, information was obtained in a series of reports, all of which were written in Japanese. The documents received from Japan and the reference which will be utilized in this text are listed below.

Report	Reference
Fukuyama-City Nakatsuhara Water Plant Sludge Treatment Operations Report (no date)	Magara 1988a
Hiroshima-City Midorii Water Works Sludge Treatment Operations Report, Feb. 26, 1988	Magara 1988b
Itami-City Senzou Water Works Sludge Treatment Facilities Preliminary Design Report, Mar. 1985	Magara 1988c
Itami-City Senzou Water Works Sludge Treatment Operations Report, Dec. 23, 1987	Magara 1988d
Saitama-Ken Water Plant Sludge Treatment Operations Report, Jan. 30, 1988	Magara 1988e
Tokyo Water Works Sludge Treatment Operations Report, Jan. 27, 1988	Magara 1988f
Yokohama-City Water Works Sludge Treatment Operations Report, Jan. 7, 1988	Magara 1988g

The documents contain basic descriptions of water treatment and sludge handling systems, as well as annual operation and performance data in tabular form. Due to differing data collection and reporting techniques used at each plant, data presentation and interpretation were highly variable requiring individual presentation of data from each site. The extent of data presentation varied by source, with some plants reporting detailed operation and performance information which is further evaluated herein. However, others reported only a brief summary of operational experiences.

The plants identified as employing coagulant recovery in Japan are presented in Table 46 along with potable and industrial water treatment capacities and type of commercial coagulants utilized. Recovered coagulants are used at all of the facilities mentioned for the production of both potable and industrial waters. Potable-water production ranges from 90,000 m^3/d (24 mgd) at Senzou to 1,300,000 m^3/d (343 mgd) at Okubu. Three of the plants, Nakatsuhara, Okubu and Kosuzume, also produce industrial water which does not have to meet stringent quality standards for potable water. Polyaluminum chloride (PACL) is the most popular commercial coagulant utilized, while alum is used exclusively at two sites and in conjunction with PACL at two other sites. All nine plants presented utilize sulfuric acid to condition sludge solids and

TABLE 46

Summary of Potable- and Industrial-Water Production and Coagulant Addition at Japanese Water Treatment Plants Practicing Coagulant Recovery (Magara 1988a-g)

Plant Name	Location	Finished Water Production, m^3/day			Commercial Coagulant
		Potable	Industrial	Total	
Nakatsuhara	Fukuyama-City	100,000	240,000	340,000	Alum
Midorii	Hiroshima-City	216,000	-	216,000	PACL*, alum
Kosuzume	Yokohama-City	-	-	1,009,200	PACL
Osaka	Tokyo	1,700,000	-	1,700,000	PACL
Misano	Tokyo	3,004,000	-	3,004,000	PACL
Higashimora-yama	Tokyo	1,265,000	-	1,265,000	PACL
Okubu	Saitama-Ken	1,300,000	250,000	1,550,000	PACL, alum
Shouwa	Saitama-Ken	350,000	-	350,000	PACL
Senzou	Itami-City	90,000	-	90,000	Alum

[1]PACL = Commercial Polyaluminum Chloride

recover coagulant for reuse at the site. The Nakatsuhara facility reuses recovered coagulant exclusively for the production of industrial water, with commercial coagulants employed for potable-water treatment. Otherwise, recovered coagulants are utilized for treatment of both potable and industrial waters at the other plants.

Information obtained on the Nakatsuhara plant contained extensive process descriptions and was found to be representative of the general water treatment and sludge handling facilities at all nine plants investigated. Therefore, the water treatment and coagulant recovery processes employed at the Nakatsuhara plant will be described in detail herein and used to represent the water and sludge treatment processes in all nine plants, with noted exceptions.

Schematic diagrams of the water treatment and sludge handling facilities are presented in Figures 51 and 52 for the Nakatsuhara plant. Pre-sedimentation is practiced prior to pre-chlorination at the pump lift station, which provides 100,000 m^3/d (26 mgd) of water for production of potable water. Commercial or recovered coagulant and lime are added in a rapid mix unit followed by mechanical flocculation and gravity sedimentation. Rapid sand filtration is employed prior to chlorine contact and distribution. On the industrial-water portion of the plant, rapid sand filtration and post-chlorination processes are not utilized due to less stringent regulatory standards on finished water quality.

Sludge solids from all sedimentation basins are combined for acid conditioning in the coagulant recovery system prior to filter press dewatering, as presented in Figure 52. The sedimentation basins on the potable-water portion of the plant have a detention time of approximately 3 h and a maximum overflow rate of 1.3 m/h (765 gpd/ft^2) at a maximum flow rate of 100,000 m^3/d. Sludge solids from the underflow of the sedimentation basin flows to primary thickening with a total surface area of 1062 m^2 where the sludge is typically thickened to approximately 7.0 percent solids. Underflow from primary thickening (0.25 m^3/min) flows to a 2.60-m^3 stainless steel acid contact basin where solids are contacted with concentrated sulfuric acid. The detention time in the acid-contact basin is about 10 min.

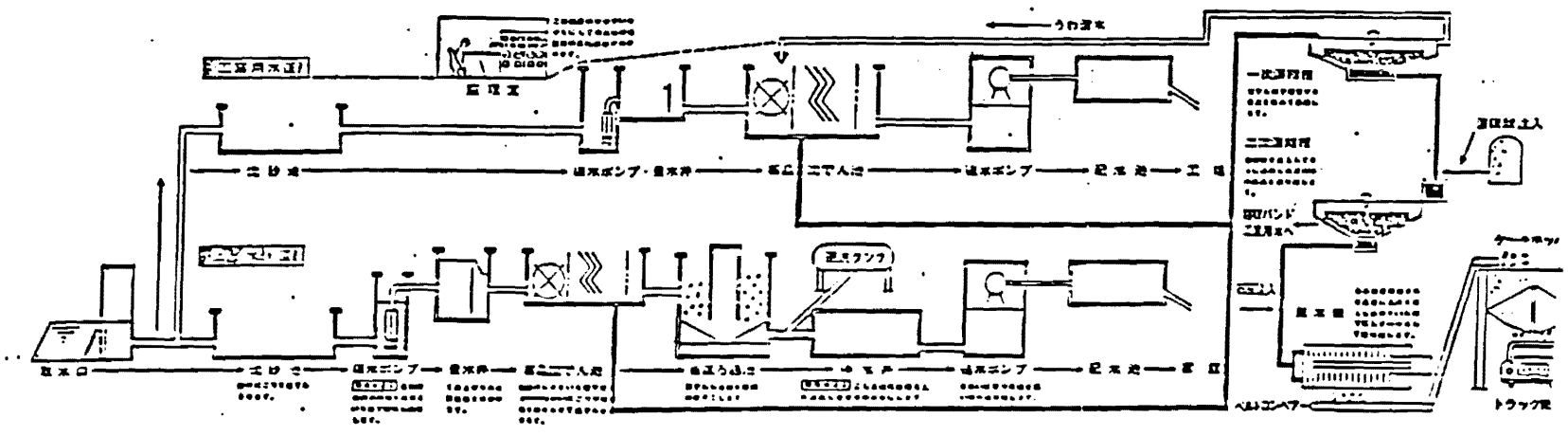

FIGURE 51. Potable and Industrial Water Treatment Flow Diagram for the Nakatsuhara Facility at Fukuyama-City in Japan (Magara 1988a)

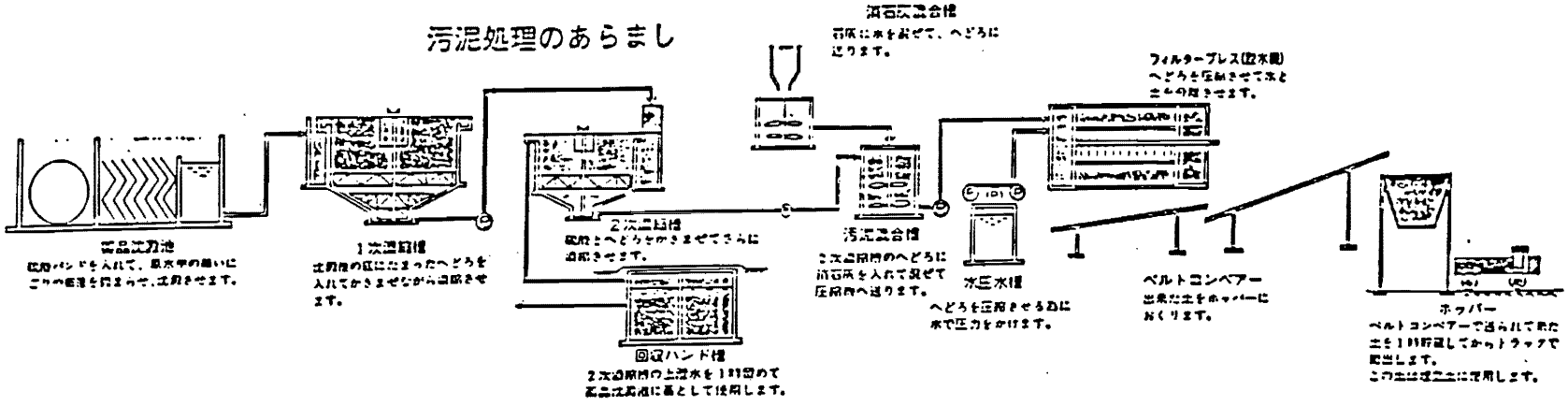

FIGURE 52. Sludge Conditioning and Coagulant Recovery Diagram for the Nakatsuhara Facility at Fukuyama-City in Japan (Magara 1988a)

Secondary thickening, with a total surface area of 133 m^2, provides separation of residual solids from the aluminum-rich supernatant liquid. Overflow from the secondary thickener is collected in a 700 m^3 (16.9m x 12.9m x 3.5m) storage tank for use as recovered coagulant. Underflow from the secondary thickeners is conditioned with lime prior to dewatering in three, 1.4-m x 1.4-m filter presses (14 chamber, 49-m^2). Cake solids from the filter press are collected in a sludge hopper and ultimately disposed of in a landfill.

Although the nine plants examined in this investigation do not have the same physical dimensions, the Nakatsuhara plant is indicative of general process schemes utilized at all of the plants. Due to the similarities in treatment processes at all nine plants, the Nakatsuhara plant will be considered to be indicative of process schematics at all nine facilities, with a few noted exceptions. Although the physical facilities of the nine plants investigated are similar, sludge characteristics (e.g., suspended solids concentration, aluminum content and pH) and coagulant recovery operations (e.g., operational pH and acid additions) vary considerably and must be examined individually.

Coagulant Recovery Operational Parameters During Full-scale Use in Japan

The information obtained from the Institute of Public Health in Japan has allowed for detailed investigation of coagulant recovery operational parameters (i.e., pH and acid addition), recovered coagulant quality (i.e., aluminum and contaminant concentrations) and recovered product utility as determined by coagulant application rates and finished water quality during full-scale operation in Japan. Reactor pH and acid addition rates have been reported for a number of the plants investigated, and are presented herein. As indicated previously, data reporting techniques varied considerably at each plant, thereby facilitating that the data be presented on a plant by plant basis. A summary of all operational data from all nine plants is also provided.

Nakatsuhara Plant in Fukuyama-City

Operational parameters for the recovery process at the Nakatsuhara plant for the period of April 1985 through March 1986 are presented in Table 47. The

TABLE 47

Monthly Operational Data for Coagulant Recovery at the Nakatsuhara Facility in Fukuyama-City for the Period April 1985 through March 1986

Month	Thickened Sludge for Acid Extraction		H_2SO_4 Usage		Recovered Coagulant		Normalized[3] Acid Dose
	Sludge Volume m^3	Suspended Solids[1] Concentration %	kg/mo	kg/kg SS	Volume m^3/mo	Al Concentration[2] mg/L	kg H_2SO_4/ kg Al Recovered
April	1694	5.5	12.275	0.132	1050	138; 259	0.41
May	1199	7.3	10.365	0.118	856	90; 111	0.12
June	1368	6.1	8.336	0.099	428	95; 74	0.23
July	1647	10.4	6.864	0.040	856	69; 58	0.13
August	1301	11.7	9.216	0.060	1226	58; 79	0.11
September	573	10.2	3.712	0.064	563	79; 69	0.09
October	958	9.4	7.680	0.085	1254	69	0.09
November	591	8.6	11.114	0.219	568	116; 127	0.16
December	281	3.8	4.972	0.272	527	185; 132	0.06
January	503	3.5	8.344	0.473	1332	111	0.06
February	721	3.2	9.078	0.393	1105	323; 344	0.02
March	604	4.7	6.824	0.219	928	206	0.04
Average	953	7.0	8.232	0.181	891	133*	0.13
Standard Deviation	476	3.0	2.452	0.139	312	84	0.11

[1]Monthly average
[2]Determined on grab sample (i.e., not monthly average)
[3]Using average grab sample aluminum concentration
*Average of all grab-sample values reported

suspended solids concentration from the primary-thickener underflow averaged 7.0 percent, ranging from 3.2 to 11.7 percent with a standard deviation of 3.0 percent. The aluminum content of the sludge was not reported.

Sludge solids are conditioned with concentrated sulfuric acid to obtain a target recovery pH value of about 3.8 in the acid-contact tank. The resulting aluminum concentration in the recovered product averaged 133 mg/L as Al, ranging from 58 to 344 mg/L as Al. Referring to the aluminum pC-pH diagram in Figure 1 at a pH value of 3.8, only approximately 270 mg/L (10^{-1} moles) of aluminum is soluble. Therefore, the above aluminum data appear to indicate that supernatant products were at or near maximum aluminum saturation. However, the data presented by other investigators (Table 12) indicates that pH values for effective aluminum extraction should be in the range of 1.0 to 3.0. Based on these results, the pH value employed in the recovery operation at the Nakatsuhara facility controlled the degree of aluminum extraction due to limited solubility of aluminum at the reported operational pH value of 3.8.

The total volume of recovered coagulant produced on a monthly basis ranged from 430 to 1250 m^3, averaging 891 ± 312 m^3 over the 12-month period. Using the reported aluminum concentration in the recovered coagulant and the volume recovered, acid addition was normalized to aluminum recovered (i.e., kg H_2SO_4/kg Al recovered) and is presented in Table 47. The normalized acid dose ranged from 0.02 to 0.41 kg H_2SO_4/kg Al recovered, with an average of 0.13 ± 0.11 kg H_2SO_4/kg Al recovered. The stoichiometric acid demand for dissolution of aluminum in the form of $Al(OH)_3 \cdot 3H_2O$ to Al^{3+} is 5.44 kg H_2SO_4/kg Al. This value is more than one order of magnitude higher than the value of 0.13 ± 0.11 kg H_2SO_4/kg Al recovered reported at the Nakatsuhara plant. Since aluminum dissolution was greater than predicted based on stoichiometric dissolution to Al^{3+}, it is reasonable to indicate that aluminum in the recovered coagulant was in the form of soluble monomers of aluminum [i.e., $Al(OH)^{2+}$ and $Al(OH)_2^{1+}$] and not exclusively in the form of Al^{3+} in the recovered solution. It is furthermore reasonable to indicate that additional aluminum could be extracted if a lower pH value (i.e., <3.8) were used.

Midorii Plant in Hiroshima-City

The Midorii facility treats 216,000 m^3/d (57 mgd) of water for potable use with polyaluminum chloride and alum as primary commercial coagulants. Coagulant recovery operational parameters are presented in Table 48. The sludge suspended solids concentration from primary thickening averaged 3.0 percent (± 0.93 percent) and was conditioned with concentrated sulfuric acid to an average pH value of about 2.0. The resulting recovered aluminum concentration averaged 1739 mg/L as Al, ranging from 1590 to 1975 mg/L as Al. The pH value of 2.0 employed at this facility is consistent with values reported in the literature (Table 12), resulting in adequate aluminum dissolution and a recovered coagulant with aluminum concentrations in a range typical of those found by numerous investigators (Tables 14 and 15).

The volume of coagulant recovered on a monthly basis ranged from 960 to 2777 m^3 ($2.56x10^5$ to $7.33x10^5$ gal). Utilizing the reported monthly acid addition and recovered aluminum concentration data, the normalized acid dose averaged 7.98 ± 0.65 kg H_2SO_4/kg Al recovered, which is about 50 percent higher than the stoichiometric dose of 5.44 kg H_2SO_4/kg Al predicted for complete dissolution of $Al(OH)_3 \cdot 3H_2O$. Since the mass of aluminum recovered is based on volumetric recovery of supernatant and its aluminum concentration, incomplete supernatant recovery (i.e., loss of aluminum associated with residual water in the sludge solids) can have a dramatic impact on the apparent acid demand in the recovery process. Comparison to acid addition data collected during full-scale testing presented earlier, i.e., 7.27 kg/kg Al by Bishop et al. (1987) and 4.1 kg/kg Al by Roberts and Roddy (1960), indicates that the average value of 7.98 kg H_2SO_4/kg Al recovered is typical of those obtained in experimental investigations.

Kosuzume Plant in Yokohama-City

The Kosuzume facility treats a total of $1x10^6$ m^3/d for potable and industrial purposes utilizing polyaluminum chloride as the primary commercial coagulant. As presented in Table 49, the suspended solids concentration to the coagulant recovery system averages 4.30 percent (± 1.78 percent), ranging from 2.6 to 9.4 percent. The total volume of recovered coagulant produced on a monthly

TABLE 48

Monthly Operational Data for Coagulant Recovery at the Midorii Facility in Hiroshima-City for the Period April 1985 through March 1986

Month	Thickened Sludge Suspended Solids Concentration %	H_2SO_4 Used kg/mo	pH	Recovered Coagulant Volume[1] m^3	Recovered Coagulant Aluminum Concentration mg/L	Relative Acid Dose[2] kg H_2SO_4/kg Al
April	3.54	17296	2.15	1408	1667	7.37
May	3.08	28152	2.03	2115	1623	8.20
June	2.34	24840	1.92	1806	1689	8.14
July	4.97	27324	1.91	1842	1716	8.64
August	3.39	44362	2.00	3241	1782	7.68
September	2.74	39761	1.93	3141	1579	8.02
October	2.47	32954	1.95	2777	1590	7.46
November	1.81	26091	1.99	2015	1755	7.38
December	1.62	21252	2.04	1612	1788	7.37
January	3.2	22816	2.06	1281	1975	9.02
February	3.9	12622	2.18	960	1782	7.38
March	2.6	25484	2.00	1458	1925	9.08
Average	3.0	26913	2.01	1971	1739	7.98
Standard Deviation	0.93	8844	-	732	123	0.65

[1]Volume of recovered coagulant used
[2]Calculated using volume of coagulant used and reported aluminum concentration

TABLE 49

Monthly Operational Data for Coagulant Recovery at the Kosuzume Facility in Yokohama-City for the Period April 1985 through March 1986

Month	Solids Concentration[1] %	H_2SO_4 Used Mg/mo*	Recovered Coagulant		Relative Acid Dose H_2SO_4/kg Al
			Volume m^3	Al Concentration[2] mg/L	
April	4.8	121.39	9962	1300	4.9
May	4.3	123.71	5238	1790	6.9
June	4.1	169.77	12718	1670	4.2
July	9.4	111.68	4684	1610	7.8
August	5.3	163.09	5879	1870	7.8
September	3.7	126.67	8262	1770	4.6
October	3.3	141.60	9423	1560	5.1
November	3.3	132.81	10605	1340	4.9
December	3.4	133.62	13635	1510	3.4
January	4.3	117.14	14185	1430	3.0
February	3.1	117.09	13785	1420	3.2
March	2.6	125.78	17452	1260	3.0
Average	4.30	132.03	10486	1544	4.90
Standard Deviation	1.78	18.07	4009	202	1.75

[1]Monthly average
[2]Assumed Sg = 1.0 for calculation of concentration

basis averaged 10486 m^3 ($2.77x10^6$ gal), ranging from 5238 to 17452 m^3 ($1.38x10^6$ to $4.61x10^6$ gal), while the aluminum content in the recovered coagulant averaged 1544 mg/L as Al (± 202 mg/L as Al). Based on the recovered volume and concentration data, the normalized acid dose was determined to be 4.90 ± 1.75 kg H_2SO_4/kg Al recovered. A recovery pH value was not specified for the Kosuzume facility.

Okubu and Shouwa Plants in Saitama-Ken

The Okubu facility treats $1.3x10^6$ m^3/d for potable water and $2.5x10^3$ m^3 of water for industrial use utilizing both polyaluminum chloride and alum as primary commercial coagulants. The Shouwa facility treats $3.5x10^5$ m^3/d for potable water utilizing polyaluminum chloride as primary commercial coagulant. Recovered coagulant is used for the treatment of drinking water at the Shouwa facility, while recovered coagulant is used for both potable- and industrial water production at the Okubu facility.

Okubu Plant. Operational data for the coagulant recovery process at the Okubu facility are presented in Table 50. Solids are thickened to about 6.7 percent (± 4.6 percent) in the primary thickener prior to acidification to a pH value of about 3.4 with concentrated sulfuric acid. The recovered coagulant aluminum concentration averaged 2276 mg/L as Al (± 650 mg/L as Al), ranging from 1430 to 3330 mg/L as Al. Sludge solids in the secondary thickener following acidification averaged 11.7 percent (± 5.6 percent), an increased suspended solids concentration of 75 percent from the primary to secondary thickeners. Normalized acid additions could not be determined due to incomplete data.

Shouwa Plant. Operational data for the coagulant recovery process at the Shouwa facility are presented in Table 51. Coagulant sludges are thickened to about 6.4 percent (± 3.1 percent) in the primary thickener prior to acidification with concentrated sulfuric acid in an acid-contact basin. The pH value in the recovery process averaged 2.5, resulting in an average recovered aluminum concentration of 2283 mg/L as Al (± 233 mg/L as Al). The secondary thickener increased the sludge solids concentration more than two-fold to about 14.2 percent (± 4.4 percent).

TABLE 50

Monthly Operational Data for Coagulant Recovery at the at the Okubu Facility in Saitama-Ken for the Period April 1985 through March 1986

Month	Suspended Solids Concentration of Thickener Underflow		Recovered Coagulant[2]		pH[1]
	Primary %	Secondary %	% Al_2O_3	mg/L as Al	
April	4.3	8.2	0.50	2650	3.39
May	4.9	10.5	0.50	2650	3.37
June	5.7	10.5	0.30	1590	3.31
July	15.9	22.1	0.27	1430	3.18
August	13.8	21.3	0.57	3020	3.27
September	12.0	16.6	0.63	3330	3.38
October	7.0	13.9	0.50	2650	3.24
November	5.6	11.0	0.39	2060	3.32
December	3.1	7.7	0.29	1530	3.55
January	2.7	6.2	0.28	1480	3.62
February	2.7	6.4	0.43	2270	3.48
March	3.2	6.5	0.50	2650	3.34
Average	6.7	11.7	0.43	2276	3.37
Standard Deviation	4.6	5.6	0.12	650	-

[1]Monthly average
[2]Calculated from %Al_2O_3 assuming suspension specific gravity of 1.0

TABLE 51

Monthly Operational Data for Coagulant Recovery at the Shouwa Facility in Saitama-Ken for the Period April 1985 through March 1986

Month	Suspended Solids Concentration of Thickener Underflow		Recovered Coagulant[2]		pH[1]
	Primary %	Secondary %	% Al_2O_3	mg/L as Al	
April	8.4	11.8	0.43	2270	2.70
May	8.3	15.2	0.48	2540	2.58
June	7.0	13.3	0.42	2220	2.48
July	13.3	21.0	0.48	2540	2.34
August	7.4	19.6	0.52	2750	2.22
September	7.0	17.0	0.42	2220	2.57
October	8.1	18.6	0.45	2380	2.38
November	4.2	16.0	0.40	2120	2.54
December	3.4	11.6	0.40	2120	2.42
January	2.6	9.3	0.36	1990	2.66
February	3.2	8.3	0.42	2220	2.45
March	3.5	8.8	0.40	2120	2.70
Average	6.4	14.2	0.43	2283	2.50
Standard Deviation	3.1	4.4	0.04	233	0.15

[1]Monthly average
[2]Calculated from %Al_2O_3 assuming suspension specific gravity of 1.0

Senzou Plant in Itami-City

The Senzou facility treats about 90,000 m^3/d of water for potable purposes employing alum as the primary commercial coagulant. Sulfuric acid additions were reported as a percent of the suspended solids concentration, or kg H_2SO_4/kg suspended solids. As presented in Table 52, acid additions from April 1983 through March 1984 averaged 0.50 kg H_2SO_4/kg SS, ranging from 0.38 to 0.65 kg H_2SO_4/kg SS. The pH value attained with the indicated dose was not reported. These monthly data were used to calculate an annual average from 1978 through 1983 as presented in Table 53. Sulfuric acid addition averaged 0.67 kg H_2SO_4/kg SS (± 0.21 kg H_2SO_4/kg SS) of the suspended solids concentration.

Summary of Coagulant Recovery Operational Parameters

A summary of acid addition data and recovery pH values employed in full-scale coagulant recovery operations in Japan is presented in Table 54. Comparison of the reported recovery pH values to those reported in experimental investigations earlier (Table 12) indicates that the pH of aluminum recovery at the Nakatsuhara facility is comparatively high, resulting in low aluminum solubility and the low aluminum concentrations in recovered coagulants. The recovery pH was also high at the Okubu facility (pH = 3.37), but resulted in recovered aluminum concentrations of about 2280 mg/L as Al. The Midorii and Shouwa recovery pH values of 2.0 and 2.5, respectively, are comparable to other pH values reported in experimental investigations in the literature.

Acid addition data indicate that the Nakatsuhara facility uses the lowest acid application rate at 0.18 kg H_2SO_4/kg SS, resulting in the relatively high pH value reported in the recovery process and the low recovered aluminum concentrations. Acid application rates reported at Midorii and Kosuzume are comparable to other full-scale experimental investigations, and indicates that more acid may be required for adequate aluminum extraction than predicted based on stoichiometric dissolution of $Al(OH)_3 \cdot 3H_2O$ to Al^{3+}.

TABLE 52

Monthly Operational Data for Coagulant Recovery at the Senzou Facility in Itami-City for the Period April 1985 through March 1986

Month	H_2SO_4 Addition[1] kg H_2SO_4/kg SS	Lime Addition[1] kg/kg SS	Total Dry Solids kg/mo	Cake Solids[1] %
April	39.6	87.0	49413	40.2
May	40.0	77.7	50173	41.9
June	37.5	76.0	44685	40.9
July	42.5	64.8	44764	39.3
August	51.9	53.6	61524	39.4
September	65.0	65.9	50061	35.0
October	56.3	61.3	27755	34.6
November	54.6	79.2	50523	33.5
December	62.1	76.4	48447	30.2
January	55.4	104.3	36449	31.6
February	44.6	77.5	47771	32.6
March	49.9	77.9	47481	34.6
Average	50.0	75.1	46587	36.2
Standard Deviation	9.1	13.1	8211	4.0

[1]Monthly average

TABLE 53

Summary of Annual Average Coagulant Recovery Operational Data at the Senzou Facility in Itami-City Between 1978 and 1983

Year	H_2SO_4 Addition kg/kg SS	Lime Addition kg/kg SS	Total Dissolved Solids kg/kg SS	Cake Solids kg/kg SS
1978	106.9	84.2	41,569	35.7
1979	70.1	79.7	53,262	35.9
1980	66.1	72.2	56,021	35.7
1981	51.8	49.4	69,986	35.6
1982	56.6	72.2	53,920	37.4
1983	50.0	75.0	46,587	36.1
Average	66.9	72.1	53,558	36.1
Standard Deviation	21.1	12.1	9,680	0.7

[1]Annual average based on monthly averaged data over the periods.

TABLE 54

Summary of Coagulant Recovery pH Values, Sulfuric Acid Additions and Recovered Coagulant Aluminum Concentrations Reported During Full-Scale Operation in Japan During 1978 to 1983

Plant	pH	H_2SO_4 Addition		Average Recovered Aluminum Concentration, mg/L as Al
		kg/kg SS	kg/kg Al Recovered	
Nakatsuhara	3.81	0.181	-	133
Midorii	2.01	-	7.98	1739
Kosuzume	-	-	4.90	1544
Okubu	3.37	-	-	2276
Shouwa	2.50	-	-	2283
Senzou	-	0.50	-	-

Comparison of recovered coagulant aluminum concentrations indicates that coagulant recovery experienced at the Nakatsuhara facility is poor, with an average of only 133 mg/L as Al compared to 1544 to 2283 mg/L as Al at the other facilities and 740 to 3700 mg/L as Al for sludges with $\geq$ 1 percent solids concentrations being typical of other coagulant recovery investigations (Tables 14 and 15).

Recovered Coagulant Aluminum and Impurity Content

Information regarding the impurity content of recovered coagulants was collected at the Kosuzume, Midorii and Okubu facilities and is reported in Table 55. A "brown, or tea-colored" product resulted from the acidification of alum sludge at all three facilities, indicating the presence of soluble organic carbon and ferric iron. Total organic carbon, however, was reported for only one recovered coagulant product at a concentration of 505 mg/L. Suspended solids and turbidity were also reported at this facility at 2400 mg/L and 12000 degree, respectively, indicating a high solids content in the recovered coagulant. The density of the recovered coagulants ranged from 1010

TABLE 55

Physical and Chemical Characteristics of Recovered Coagulants Produced at the Kosuzume, Midorii and Okubu Facilities

	Kosuzume		Midorii				Okubu
Sample	#1	#2	#1	#2	#3	#4	
pH	2.2	2.6	2.0	2.0	2.0	1.9	3.4
Density, g/mL	1.014	1.010	1.019	1.012	1.013	1.020	-
Aluminum	1877	1336	2264	1445	1715	2482	-
Turbidity, degree	-	-	-	-	-	-	12000
Suspended Solids, mg/L	-	-	-	-	-	-	2140
Color	brown	brown	brown	brown	brown	brown	brown
TOC, mg/L	-	-	-	-	-	-	505
NH_4-N, mg/L	30	15	-	-	-	-	2.52
Arsenic, mg/L	0.0	0.0	2.6	1.3	1.2	1.8	0.143
Cadmium, mg/L	0.0	0.0	0.005	0.004	0.005	0.008	0.011
Copper, mg/L	-	-	0.79	0.21	0.52	0.41	1.15
Chromium, mg/L	0.0	0.0	0.00	0.00	0.00	0.00	0.02
Iron, mg/L	67	18	224	233	263	439	210
Lead, mg/L	0	0	0.37	0.32	0.32	0.66	0.07
Manganese, mg/L	62	50	17.8	10.2	11.0	19.6	60
Mercury, mg/L	0.00	0.00	0.002	0.001	0.001	0.000	0.0008
Zinc, mg/L	-	-	-	-	-	-	31

kg/m^3 at the Kosuzume facility (sample #2) to 1020 kg/m^3 at the Midorii facility (sample #4). The average pH of recovered solutions varied from 1.9 to 3.8 among the facilities, but was fairly consistent at each plant.

The total aluminum content in the recovered coagulants ranged from 1336 to 2482 mg/L at the three facilities reported in Table 55, and was considerably lower at an average of 133 mg/L at the Nakatsuhara facility. As discussed previously, the low recovered coagulant aluminum concentration at the Nakatsuhara facility was the result of low acid application rates and the relatively high pH value utilized in the recovery process.

Following aluminum, iron was the most predominant metal present in the recovered solutions, at concentrations ranging from 18 to 439 mg/L as Fe. Manganese concentrations were also high, ranging from 10.2 to 62 mg/L in the recovered coagulants. Zinc was determined to be 31 mg/L at the Okubu facility, while it was not measured in the other recovered coagulants.

Arsenic was the next largest constituent in the recovered coagulant products ranging from zero to 2.6 mg/L. Copper concentrations ranged from 0.21 to 1.15 mg/L, while lead concentrations ranged from zero to 0.66 mg/L in the recovered coagulants. Other heavy metals, such as mercury, cadmium and chromium, were all reported at significantly lower concentrations ranging up to 0.02 mg/L.

Heavy metal concentrations were normalized to recovered aluminum concentrations (i.e., mg/kg Al recovered) for comparison to the Water Chemicals Codex (National Research Council 1982) and are presented in Table 56. It should be noted that the concentrations reported were not determined using standard Water Chemicals Codex (National Research Council 1982) procedures. Therefore, as discussed previously, this comparison is conservative in that it does not exclude the fraction of the contaminants which do not remain in the filtered solution at a pH value of 6.0. Iron is not regulated by the Water Chemicals Codex (National Research Council 1982) and was measured in the recovered coagulants at normalized concentrations ranging from 35,695 mg/kg recovered Al to 176,875 mg/kg recovered Al. Normalized manganese concentrations were also high, ranging from 6,414 to 37,425 mg/kg recovered Al, while copper ranged from 145 to 349 mg/kg Al

TABLE 56

Normalized Metal Concentrations (mg/kg Al) in Recovered Coagulants Produced at the Kosuzume and Midorii Facilities

	Impurity Concentration, mg/kg Al						
	Kosuzume		Midorii				RMIC[1]
Metal	#1	#2	#1	#2	#3	#4	mg/kg Al
Arsenic	0.0	0.0	1148	900	700	725	330
Cadmium	0.0	0.0	2.2	2.8	2.9	3.2	77
Copper	-	-	349	145	303	165	-
Chromium	0.0	0.0	0.00	0.00	0.00	0.00	330
Iron	35695	13473	98940	161246	153353	176873	-
Lead	0	0	163	221	187	266	330
Manganese	33031	37425	7862	7059	6414	7897	-
Mercury	0.00	0.00	0.9	-	-	-	11
Zinc	-	-	-	-	-	-	-

[1]Recommended Maximum Impurity Content specified by the Water Chemicals Codex (National Research Council 1982) using a maximum alum dose of 150 mg/L and a safety factor of 10.

recovered. Manganese and copper concentrations are not regulated by the Water Chemicals Codex (National Research Council 1982).

With the exception the Kosuzume facility, the normalized arsenic concentrations exceeded the RMIC value of 330 mg/kg Al in all cases by more than two-fold. Normalized lead concentrations were also notably high at concentrations ranging from 163 to 266 mg/kg recovered Al, but did not exceed the RMIC value of 330 mg/kg Al. All other contaminant concentrations (i.e., cadmium, chromium and mercury) were more than one order of magnitude lower than RMIC value established by the Water Chemical Codex (National Research Council 1982).

Recovered Coagulant Utility and Finished Water Quality

Recovered coagulant application rates were reported for the Senzou, Kosuzume and Midorii plants. Mass application rates for commercial and recovered coagulants were compared where possible to determine the relative use of each coagulant. Finished water quality at the Senzou plant was found to be representative of finished water quality during use of recovered coagulants and will be compared to Japanese maximum contaminant levels.

Recovered Coagulant Utility

As presented earlier in Table 46, the Senzou plant uses alum as the primary coagulant to treat 90,000 m^3/d of potable water. This results in about $2x10^6$ m^3/mo of potable water being produced from the simultaneous addition of commercial and recovered coagulants in the rapid mix unit. As presented in Table 57, commercial application rates ranged from approximately 53 to 122 metric tonne/mo, resulting in average monthly aluminum doses ranging from 15.6 to 34.5 mg/L as alum (1.4 to 3.1 mg/L as Al). The total volume of recovered coagulant applied on a monthly basis ranged from 1450 to 4660 m^3/mo and volumetric application rates ranged from 0.79 to 2.00 mL/L. Using the average recovered coagulant aluminum concentration of approximately 2650 mg/L as Al (0.5 percent as Al_2O_3), the recovered coagulant application rates were determined to be equivalent to 2.1 to 5.3 mg/L as Al. Based on a comparison of commercial and recovered coagulant aluminum application rates, recovered

TABLE 57

Application Rates for Commercial and Recovered Coagulants as Added Simultaneously at the Senzou Facility in 1983-1984

Month	Water Treated m^3/mo	Commercial Alum		Recovered Coagulant	
		kg/mo	mg/L	m^3/mo	mL/L
April	1,835,670	58,490	18.21	3527.6	1.92
May	1,993,300	64,476	18.48	3464.6	1.74
June	2,044,150	81,305	22.73	3870.3	1.89
July	2,259,480	122,956	31.10	3482.0	1.54
August	2,358,720	88,887	21.53	4661.0	1.98
September	2,151,480	110,389	29.32	4308.6	2.00
October	2,055,550	95,938	26.67	1901.4	0.92
November	1,894,960	58,942	17.77	3586.7	1.89
December	1,947,120	53,293	15.64	3575.6	1.84
January	1,837,460	60,356	18.77	1446.5	0.79
February	1,806,480	53,826	17.03	1763.6	0.98
March	1,869,800	58,267	17.81	1837.2	0.98

coagulant application rates are typically equivalent to or greater than commercial application rates at this facility.

The Kosuzume water treatment plant treats an average of $1x10^6$ m^3/d of potable and industrial water utilizing polyaluminum chloride (PACL) as the primary commercial coagulant. As presented in Table 58, PACL coagulants were applied at 22.2 to 337.8 m^3/mo, resulting in volumetric application rates ranging from 9.99 to 23.2 mL/m^3. Using an estimated aluminum concentration of 50 g/L in the PACL commercial coagulants, the mass application rate of PACL ranged from 0.5 to 1.2 mg/L as Al. Recovered coagulant volumes applied on a monthly basis ranged from approximately 4.5 to 14.5 m^3/mo, resulting in volumetric application rates of 0.265 to 0.911 mL/L. Using the average aluminum concentration in the recovered coagulants of 1544 mg/L as Al, the mass application rates for recovered coagulants ranged from 0.4 to 1.4 mg/L as Al. Therefore, recovered coagulant mass application rates were equal to mass application rates for PACL.

The Midorii plant treats approximately $2.16x10^5$ m^3/d of potable water utilizing polyaluminum chloride and alum as commercial coagulants. The volume of water treated on a monthly basis is presented in Table 59 along with the recovered coagulant volume and volumetric application rate. The volume of recovered coagulant applied ranged from 960 to 3241 m^3/mo, resulting in volumetric application rates ranging from 327 to 716 mL/m^3. Using the average recovered coagulant aluminum concentration of 1739 mg/L as Al, recovered coagulant mass application rates ranged from 0.6 to 1.2 mg/L as Al. The mass application rates for PACL and alum could not be determined.

As indicated by data presented above, recovered coagulant application rates are typically on the order of 50 percent of the total coagulant aluminum mass application rate. Since the recovered coagulants are at least an order of magnitude lower in aluminum concentration as compared to commercial coagulant, a considerably larger volume of recovered coagulant is required to attain this rate. That is, when the recovered coagulant aluminum concentration is one order of magnitude less than commercial coagulant and they are added at the same mass application rate, the volumetric application of recovered coagulant is one order of magnitude larger than that for commercial coagulant. To

TABLE 58

Average Monthly Application Rates for Commercial and Recovered Coagulants as Added Simultaneously at the Kosuzume Facility in 1985-1986

Month	Polyaluminum Chloride			Recovered Coagulant	
	m^3/mo	mL/m^3	mg/L as Al	m^3/mo	mL/L
April	121.92	12.60	0.63	9,470	0.625
May	187.98	12.57	0.63	4,530	0.361
June	115.20	16.58	0.83	8,030	0.453
July	337.76	23.15	1.16	8,890	0.742
August	104.26	12.33	0.62	4,890	0.265
September	64.43	10.63	0.53	7,510	0.395
October	28.89	13.15	0.66	6,820	0.331
November	22.22	10.29	0.52	14,460	0.667
December	73.07	9.99	0.50	12,780	0.713
January	72.83	11.73	0.59	16,130	0.911
February	63.56	11.12	0.56	13,410	0.730
March	107.60	15.59	0.78	14,340	0.784

*Assumed Commercial PACL Concentration of 50 g/L as Al

TABLE 59

Monthly Average Applications Rates for Recovered Coagulants as Added Simultaneously at the Midorii Facility in 1985-1986

Month	Water Treated m^3/mo	Recovered Coagulant Use	
		Volume Applied m^3/mo	Volumetric Application Rate mL/L
April	4,266,790	1408.38	0.333
May	4,708,630	2114.88	0.472
June	4,880,160	1805.67	0.522
July	4,666,550	1841.75	0.423
August	4,869,570	3241.27	0.646
September	4,530,520	3140.91	0.716
October	4,584,490	2777.35	0.560
November	4,190,750	2014.70	0.557
December	4,333,230	1611.66	0.411
January	3,988,030	1280.62	0.426
February	3,768,040	960.10	0.387
March	4,249,880	1457.68	0.327

maintain a storage capacity for the average 30-d volume of recovered coagulant, storage tank of 2000 m^3, 3120 m^3 and 10,100 m^3 ($5.3x10^5$, $8.24x10^5$ and $2.67x10^6$ gal) would be required at the Midorii, Senzou and Kosuzume plants, respectively. Related to the stated plant capacities in Table 46, the recovered coagulant storage volumes would be from 1 to 3.5 percent of average daily plant capacity. When compared in a similar manner, the commercial coagulant storage capacities at the three plants range from 0.004 to 0.01 percent of average daily flow. Coagulant storage facilities for recovered coagulants must therefore be significantly greater than those for commercial coagulants.

Finished Water Quality

Finished water quality at the Senzou water treatment plant was found to be representative of finished water quality data at all of the plants examined. Furthermore, extensive use of the recovered coagulants for the production of potable water has been documented at this site. As discussed previously, commercial and recovered coagulants are added simultaneously at application rates ranging from 1.4 to 2.8 mg/L as Al for commercial coagulant and 2.1 to 5.3 mg/L as Al for recovered coagulant. Monthly averaged water quality data for 1983 and reported maximum contaminant levels are presented in Table 60. The pH value of the finished water ranged from 6.7 to 7.0, averaging 6.8 during 1983. Total dissolved solids averaged 163 mg/L during the period, while the average total hardness was 59.0 mg/L as $CaCO_3$. Iron and manganese concentrations were well below the maximum contaminant level of 0.3 mg/L, averaging 0.02 mg/L and zero, respectively. Other heavy metal contaminants were at least one order of magnitude below the maximum contaminant levels, while coliform levels (MPN) were found to be zero in all cases. In general, excellent finished water quality was obtained in 1983 while recovered coagulants were being used extensively, with PACL and alum being added as make-up coagulants to ensure acceptable finished water quality.

Treatment of Residual Sludge Solids Following Acidification

All of the nine plants investigated utilize filter press dewatering systems for final dewatering following sludge acidification and residual sludge solids

TABLE 60

Monthly-Average Data for Finished Water Quality at the Senzou Water Treatment Plant in 1985

	Japan MCL	Apr	May	June	July	Aug	Sept	Oct	Nov	Dec	Jan	Feb	Mar	Annual* Average
Water Temperature, °C	-	15.7	20.9	21.3	24.4	28.7	22.2	18.4	13.0	9.2	4.9	7.1	6.7	160
pH	-	6.8	6.9	6.8	6.8	6.8	7.0	6.9	6.8	6.8	6.8	6.7	6.7	6.8
Turbidity, degree	-	0	0	0	0	0	0	0	0	0	0	0	0	0
Total Alkalinity, mg/L as $CaCO_3$	-	22	23	21	20	28	28	26	31	31	30	29	25	26
Total Dissolved Solids, mg/L	500	137	156	108	124	156	160	135	192	193	230	188	168	163
Hardness														
Total, mg/L as $CaCO_3$	300	50.7	54.0	43.0	39.4	57.4	53.4	50.2	68.3	81.0	77.8	68.3	63.9	59.0
Ca, mg/L as $CaCO_3$	-	41.3	43.6	31.0	29.9	47.8	44.7	39.7	57.2	64.8	64.2	55.9	52.7	47.7
Mg, mg/L as $CaCO_3$	-	9.4	10.4	12.0	9.5	9.6	8.7	10.5	11.1	16.2	13.6	12.4	11.2	11.2
Total Iron, mg/L	0.3	0.01	0.04	0.01	0.01	0.02	0.01	0.02	0.01	0.03	0.03	0.03	0.01	0.02
Total Manganese, mg/L	0.3	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00
Copper, mg/L	1.0	0.003	0.003	0.004	0.002	0.004	0.003	0.004	0.002	0.003	0.005	0.008	0.004	0.004
Lead, mg/L	0.1	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Zinc, mg/L	1.0	0.006	0.001	0.003	0.008	0.008	0.007	0.012	0.005	0.005	0.010	0.000	0.003	0.006
Cadmium, mg/L	0.01	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Chromium, mg/L	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Arsenic, mg/L	0.05	0.000	0.000	0.003	0.000	0.004	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001
Coliform, #/100 mL	-	0	0	0	0	0	0	0	0	0	0	0	0	0

*Calculated from monthly averages

conditioning. All plants utilize lime for pH adjustment and sludge conditioning prior to application to the filter press, while the three Tokyo plants utilize lime and polymer conditioning of acidified sludge solids. Lime application rates and final sludge solids content were reported at the Senzou facility. Limited information on lime application rates and final sludge solids content was available for the other plants.

Lime application rates and final sludge solids content for the Senzou facility are presented in Table 52. Monthly and annual lime application rates ranged from 0.54 to 1.04 kg/kg SS between April 1983 and March 1984, averaging 0.75 kg/kg SS. Annual lime application rates over the period 1978 through 1984 averaged 0.72 kg/kg SS, ranging from 0.49 kg/kg SS in 1981 to 0.84 kg/kg SS in 1978. These data indicate the elevated levels of lime required to treat acidified sludges.

Following lime conditioning, the total dry solids for ultimate disposal averaged 46,587 kg/mo for 1983 and 53,558 kg/mo between 1978 and 1984. Using the average plant flow rate of 2.3×10^6 m^3/mo, the total solids for ultimate disposal normalized to plant flow was 0.023 kg/m^3 of water treated. The resulting cake solids concentrations during 1983 ranged from 30.2 to 41.9 percent, averaging 36.2 percent solids. The annual averaged cake solids concentrations (1978-1984) averaged 36.1 percent solids, ranging from 35.6 percent in 1981 to 37.4 percent in 1982.

FULL-SCALE IRON RECOVERY AT ATHENS, TENNESSEE

Plant Description and Operational Procedures

The water treatment plant at Athens, TN is a small conventional plant operated by the Athens Utility Board and providing potable water for the City of Athens, with supplemental water being supplied from the Hiwassee Utility Company. The Athens facility uses ferric sulfate as a primary coagulant and is the only full-scale operational plant using a coagulant recovery and reuse system in the United States. Recovery of the primary iron coagulant using sulfuric acid conditioning of sludge solids has been practiced since October 1985. The objective of this study presented herein was to evaluate data

provided by plant personnel (Camp 1989) to investigate the full-scale coagulant recovery system employed; to evaluate the impact of coagulant recovery on chemical additions, process operations and finished water quality; and to evaluate the economical implications of the coagulant recovery process.

Physical Facilities

Water Treatment. A schematic diagram of the water treatment system and sludge handling facility is presented in Figure 53. To initiate treatment of the raw water, ferric sulfate [$Fe_2(SO_4)_3 \cdot 5H_2O$], hydrated lime [$Ca(OH)_2$], powdered activated carbon and chlorine are added in the rapid mix tank, which has a detention time of approximately 2 min. The flocculation basin, with a retention time of 50 min, and using a paddle-wheel mixer, is followed by sedimentation basins with a retention time of about 6 h. Rapid sand filters are operated at a flow rate of 4.9 m/h (2 gpm/ft^2) and are typically backwashed with a maximum volume of about 280 m^3 (75,000 gal) of water at a rate of 46.5 m/h (19 gpm/ft^2). Head loss, as opposed to turbidity breakthrough, is typically the controlling factor in the length of filtration runs, which vary from about 60 to 120 h prior to backwashing. Finally, chlorination prior to storage and distribution is employed for disinfection.

Sludge Treatment. The sludge handling facility, as presented in Figure 53, can be operated using one of two options available for treatment of sludge solids. Sludge solids are removed from the sedimentation basins on a batch basis and can be conditioned with organic polymer and dewatered directly on a vacuum-assisted drying bed. A second option utilizes acid and polymer conditioning of sludge solids for iron dissolution and recovery for reuse as a coagulant. In both options, a vacuum-assisted drying bed with a total area of 92 m^2 (994 ft^2) is employed for dewatering sludge solids prior to ultimate disposal at a landfill.

Full-scale iron recovery has been practiced at the Athens, TN water treatment facility since October 1985 at the discretion of the operator. Sulfuric acid is added in-line to the sedimentation basin underflow as it flows to a 4.2-m^3 (1100-gal) acid-contact basin. The pH of the acid-contact basin effluent is used as the control parameter for the recovery process. The detention time in

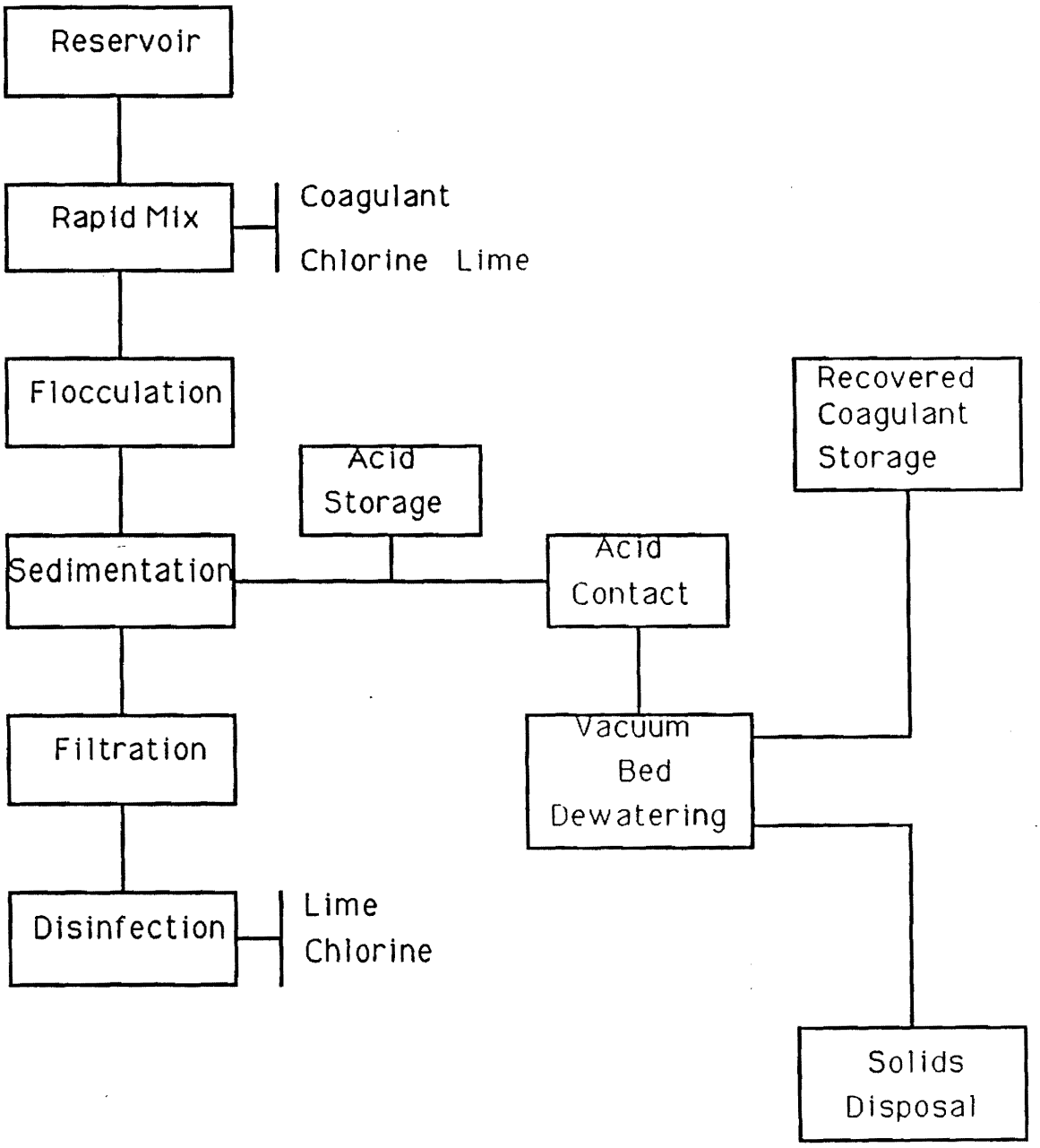

FIGURE 53. Schematic Diagram of Water and Sludge Treatment Facilities Utilized for Full-Scale Iron Recovery at Athens, TN

the acid-contact basin is dependent on flow rate and ranges from 10 to 20 min. A nonionic polymer is used for conditioning acid-extracted sludge immediately prior to application to the vacuum-assisted drying bed. Filtrate from the drying beds is collected as the recovered coagulant and stored in an 82-m^3 (21,600-gal) tank for use at the site. At the plant, the term "filtrate" is used to describe the recovered coagulant.

Acid and polymer addition rates, pH of acid extracted sludge suspension and volume of "filtrate", or recovered coagulant, produced are recorded for each recovery run. The acid addition rate is based on maintaining a target pH value, which has varied between 1.3 and 2.0. The pH of the recovered coagulant solution is also determined following storage to document any changes with time. The total volume of acid used to produce a given batch is determined from volumetric changes in the acid storage reservoir and converted to mass applied. Nonionic organic polymer is added prior to sludge application on the drying bed to aid the dewatering process. The total quantities of acid and polymer applied are reported for each run and used in an economical analysis conducted by plant personnel for each run. Manganese concentrations and pH were measured for all batches of recovered coagulant, while iron concentrations were determined on all batches since December, 1987.

Plant Operational Procedures

The Athens, TN water treatment facility is typically operated on a 5 d/wk basis, producing about 9500 m^3 ($2.5x10^6$ gal) of potable water on a daily basis. Furthermore, plant operations are semi-continuous in that the daily operational period is 17 hours and plant operations are then stopped and remain idle the remainder of the day. Daily operations are initiated on each weekday morning upon the initiation of influent flow. No special procedures are used in the initiation or termination of treatment units as a result of the semi-continuous operational mode. Furthermore, plant filters are not backwashed at the termination of service or prior to initiation of operation, unless backwashing is warranted by headloss or effluent quality considerations.

Raw Water Characteristics. The raw water for the plant is obtained from a

surface reservoir. Raw water characteristics, such as turbidity, alkalinity, pH and temperature, can have dramatic impacts on, for example, chemical application rates and operational performance. These effects are addressed herein. The 1988 raw water data are presented herein since much of the following discussion will address operations during this period due to the intensive use of the coagulant recovery system. Furthermore, the raw water data presented for 1988 are indicative of data collected since October 1985 and show a typical annual cycle of raw water quality. It should be noted that all data are plotted to show the semi-continuous nature of treatment operations. That is, the data are plotted on the date recorded. Points on successive week days are connected, while blank spaces are indicative of weekend breaks in plant operations.

Turbidity of the raw water is typically low, varying from 5 to 20 NTU, with periodic fluctuations to values approaching 1000 NTU during periods of severe weather or reservoir instability. As shown in Figure 54, during several periods in January, February and March 1988, influent turbidity increased to values between 100 NTU and 1000 NTU for 2- to 5-day periods.

Raw water alkalinity, presented in Figure 55, exhibits minimal daily variation, and is typically in the range of 135 to 160 mg/L as $CaCO_3$. While variations in alkalinity are small, it appears to increase gradually from an average of 141 mg/L as $CaCO_3$ in January 1988 to 159 mg/L in December 1988. During periods of poor raw-water quality, alkalinity is dramatically impacted and can drop to well below 100 mg/L. This is apparent when comparing data in Figures 54 and 55. The decline in alkalinity is not as dramatic as the increase in turbidity, nor does it occur with the same frequency. However, when alkalinity decreases, it is always in conjunction with an increase in turbidity.

There is a limited variation in the pH of the raw water, with typical values of 7.5 to 8.1, as presented in Figure 55. The temperature of the raw water varied seasonally, as presented in Figure 56, with values ranging from 12°C in January to 21°C in July.

Due to the use of a surface reservoir as a raw water supply, smaller

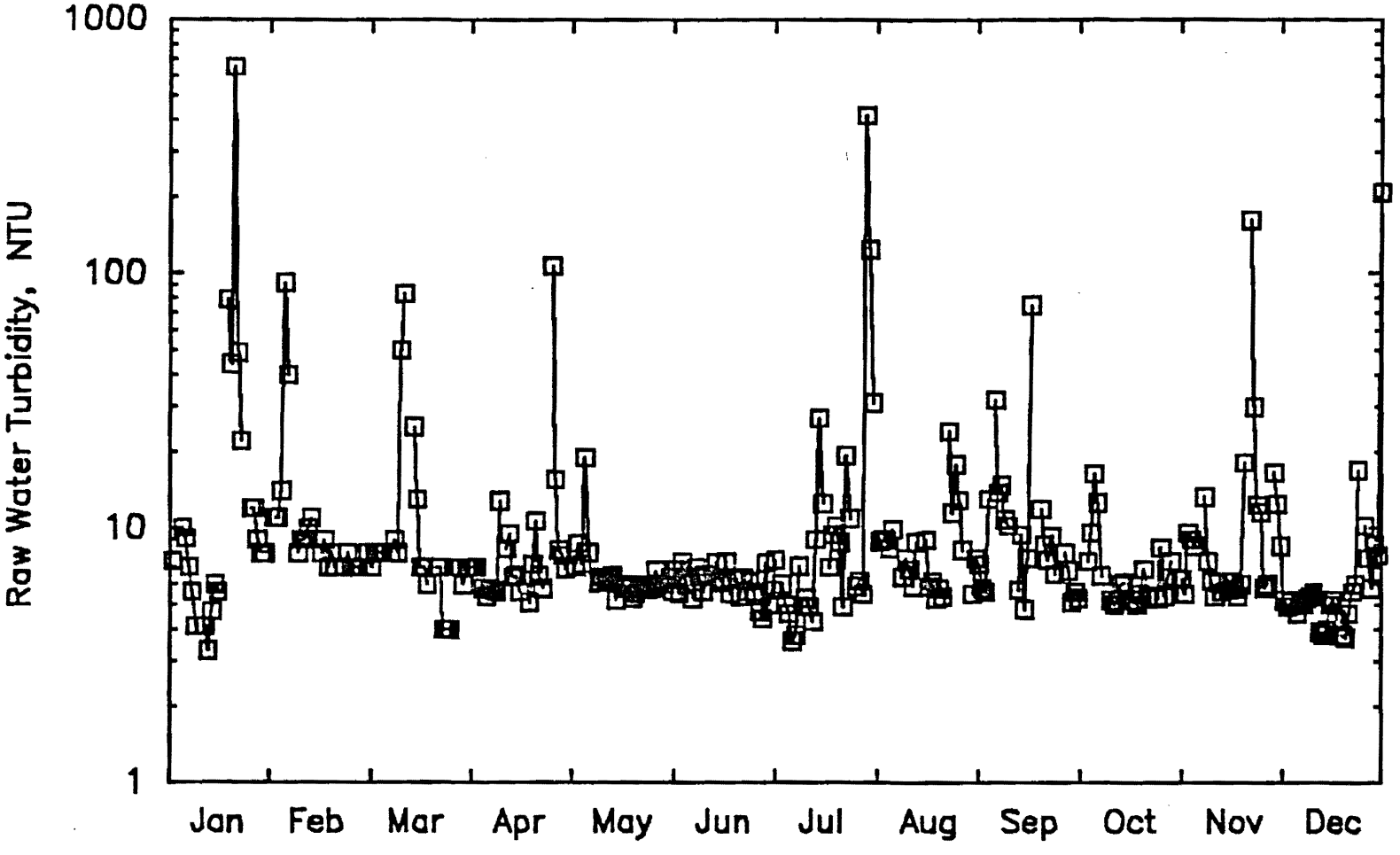

FIGURE 54. 1988 Raw Water Turbidity (log-scale) at the Athens, TN Water Treatment Plant

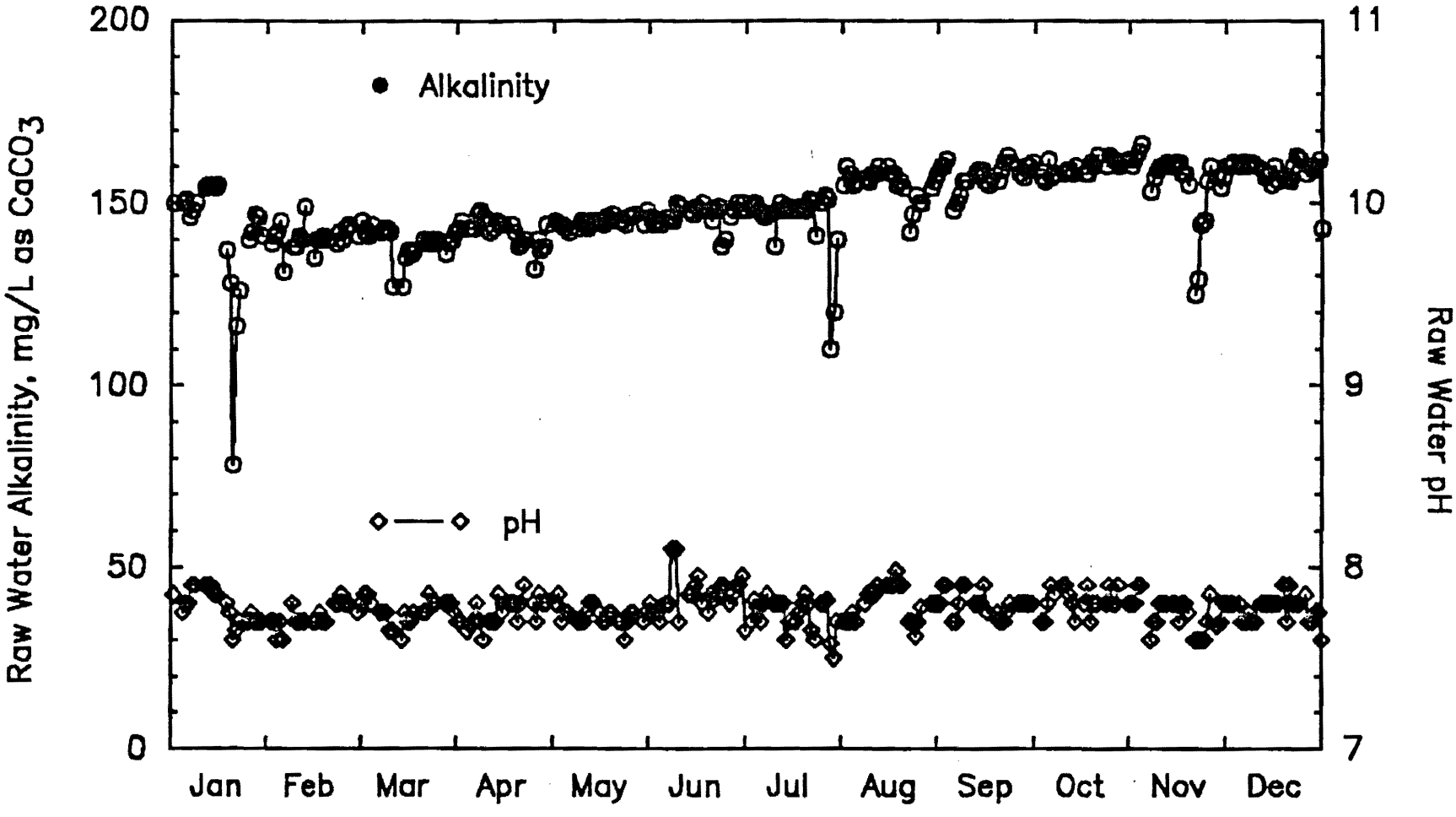

FIGURE 55. 1988 Raw Water Alkalinity and pH at the Athens, TN Water Treatment Plant

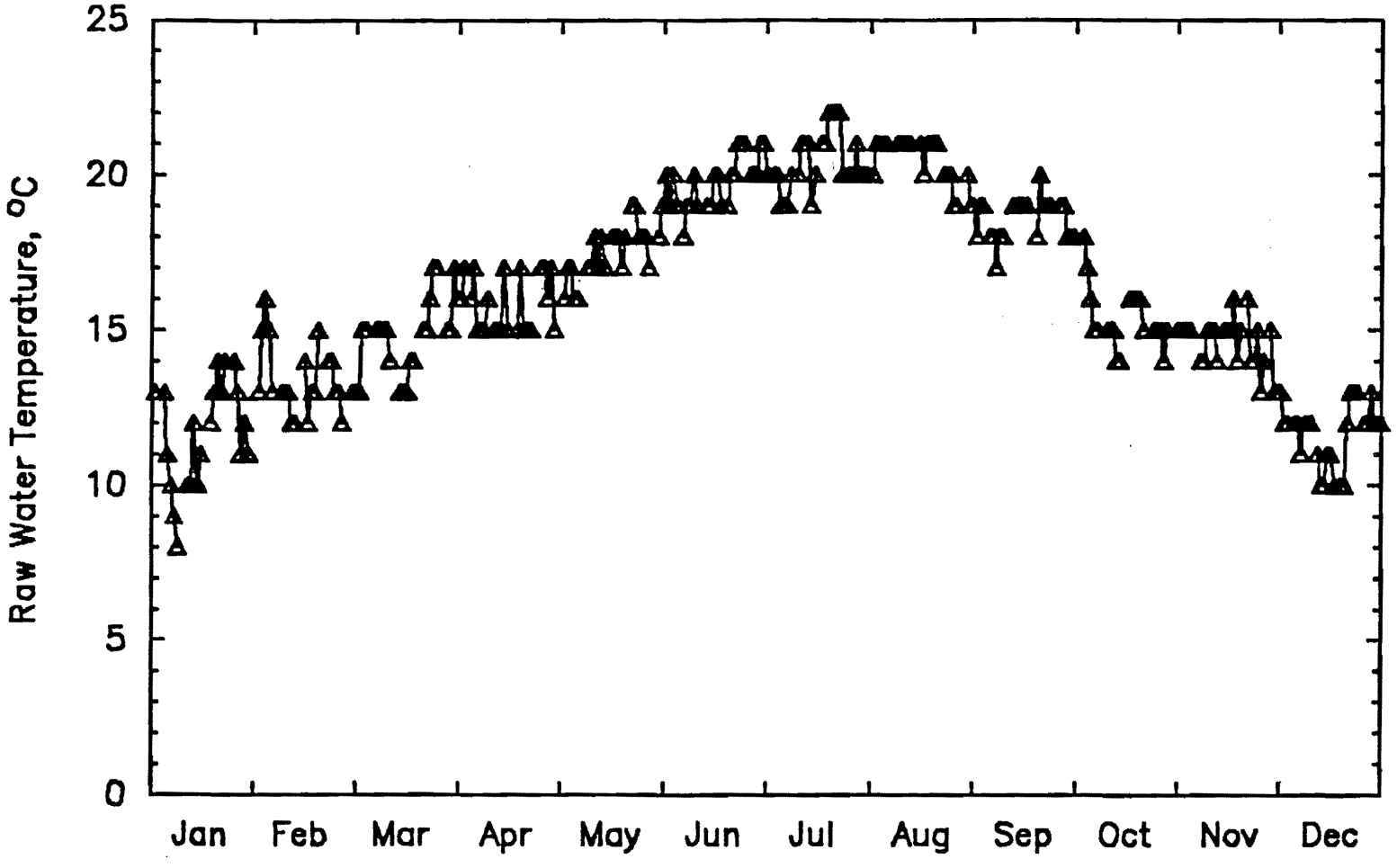

FIGURE 56. 1988 Raw Water Temperature at the Athens, TN Water Treatment Plant

variations in raw water characteristics might be expected than in those found from a river or stream source. In general, only minor variations in raw water characteristics were observed over the period of study. Furthermore, those variations encountered were seasonal variations in temperature and some extreme daily variations in turbidity and alkalinity.

Chemical Additions. A commercial solution of ferric sulfate [$Fe_2(SO_4)_3 \cdot 5H_2O$] is used as the primary coagulant and is applied at the rapid mix unit. Pertinent technical data for the commercial ferric sulfate product are presented in Table 61. The commercial product contains soluble ferric iron at a concentration of 10.0 to 10.5 percent, while soluble ferrous iron is present at much lower concentrations, ranging from 0.5 to 0.75 percent. Using a specific gravity of 1.5, this corresponds to a total iron concentration of 158 to 169 g/L as Fe.

TABLE 61

Technical Information on Liquid Ferric Sulfate Supplied by Tennessee Chemical Company (1984)

Parameter	Concentration
Soluble Ferric Iron	10.0-10.5 g Fe^{+++}/100 g solution
Soluble Ferrous Iron	0.5-0.75 g Fe^{++}/100 g solution
Free Sulfuric Acid	1.0 g H_2SO_4/100 g solution
Water Insolubles	<0.1 g/100 g solution
Density	1.48-1.50 g/mL solution

Coagulant application rates are reported by plant personnel as ferric sulfate [i.e., mg/L as $Fe_2(SO_4)_3 \cdot 5H_2O$], but are reported herein as total iron (i.e., mg/L as Fe) for purposes of comparison with other coagulant data, as described later. For reference purposes, the conversion factor for the iron content of the coagulant solution is 0.228 g Fe/g $Fe_2(SO_4)_3 \cdot 5H_2O$. Typical coagulant doses ranged from 4 to 10 mg/L as Fe [17.5 to 43.9 mg/L as $Fe_2(SO_4)_3 \cdot 5H_2O$] during periods of typical raw water quality, with periods of severe fluctuations in

doses to as high as 31 mg/L as Fe during periods of decreased raw water quality.

Lime is employed for pH and alkalinity adjustment in the rapid mix unit and is typically added at concentrations ranging from 15 to 40 mg/L to maintain the pH at a value of about 7.5 to 8.0. Similar to the coagulant dose, periods of decreased raw water quality have a significant impact on the addition of lime. That is, as ferric sulfate doses increase during periods of increased turbidity, lime doses increased to nearly 80 mg/L.

Pre-chlorination and post-chlorination are also practiced utilizing chlorine gas for disinfection and oxidation of reduced metals, e.g., Fe^{+2} and Mn^{+2}. Typical pre-chlorination doses range from 1.7 to 3.2 mg/L as Cl_2, while post-chlorination doses range from about 1.6 to 2.9 mg/L as Cl_2.

Sludge Treatment Operations. Sludge solids are removed from sedimentation basins on a batch basis. Since the sedimentation basins do not have manual or automatic sludge scrappers, underflow valves from the basins are opened and an operator manually induces breakup and flow of the sludge blanket to develop an adequate flow of concentrated sludge to the solids treatment facility using an air lance. Consequently, the suspended solids concentration in the underflow is highly variable and is not typically monitored by plant personnel.

Sludge solids can be acid and polymer conditioned in-line as they flow to a contact tank at the drying bed facility. While the coagulant recovery system is in operation, sludge solids are conditioned in-line with sulfuric acid prior to discharge to the acid-contact basin, which has a hydraulic retention time of 10 to 20 min depending on flow rate. Overflow from the acid-contact basin is treated with polymer and applied to the vacuum-assisted drying bed. During periods when coagulant recovery is not being practiced and sulfuric acid is not being added, sludge solids are polymer conditioned immediately prior to application to the drying bed, bypassing the acid-contact basin.

The coagulant recovery system has been operational since 1985 and has required several modifications by plant operators to refine the process and address problems associated with use of the highly acidic recovered coagulant product.

The configuration of the system currently employed was finalized in December 1987. Through December 1987, 10 successful coagulant recovery runs were conducted at the facility, while 19 runs were conducted during 1988. With the exception of one volume of sludge which was not acid conditioned, all sludge solids removed from the sedimentation basins in 1988 were acid and polymer conditioned. One batch of the resulting recovered coagulant was wasted due to operational problems associated with its reuse. Otherwise, all recovered coagulant produced (i.e., 18 batches) in the period of January through December 1988 was reused at the site to produce potable water.

Recovered Coagulant Production and Quality

Production of Recovered Coagulant

A total of 29 batch coagulant-recovery runs have been conducted at the Athens facility since October 1985. The total volume of recovered coagulant for a "batch" run is frequently produced over a period of several days. All recovered coagulant produced over such periods was accumulated in the 81.8 m^3 (21,600-gal) storage tank and referred to as a single batch by plant personnel. Chemical applications were typically reported as a cumulative value for the entire batch produced over several days, as opposed to individual values for each day of operation.

Sulfuric acid and polymer addition data, along with pH and volume of the recovered coagulant are presented in Table 62. The volume of recovered coagulant produced in a batch ranged from about 50 to 264 m^3 (13,000 to 70,000 gal) and was a function of the volume of sludge treated, sludge suspended solids concentration, acid and polymer doses and the dewaterability of the residual sludge solids. In many instances, the volume recovered in one batch was larger than the storage volume. However, a batch of recovered coagulant was typically produced over several days and used at the plant during the production period, thereby accounting for the discrepancy in storage volume and volume produced. The lack of information on sludge solids concentration and total volume or mass of sludge solids treated did not allow for detailed investigation of acid and polymer application rates. However, as mentioned earlier, acid addition was controlled by the pH in the reactor, which ranged

TABLE 62

Summary of Recovered Coagulant Production Data from Full-Scale Operation at Athens, TN over the Period of 1985 through 1988

Dates	Recovered Volume, m^3	Acid-Contact Tank pH	H_2SO_4 Applied, kg	Polymer		Normalized Acid Dose kg/kg Fe recovered
				Volume L	Dose ppm	
1985						
10/7, 8	>68.1	1.3-1.4	3.14	7.6	<111	-
11/1, 5, 12	127.7	2.0	3.14	18.9	148	-
12/4, 5, 17, 30	197.4	1.3-1.5	2.35	18.9	96	-
1986						
10/30, 11/10	217.6	1.9	2.99	7.6	35	-
1987						
1/6, 7, 14	-	1.8	2.73	8.5	-	-
3/4, 5, 25	227.2	1.7-1.8	4.30	11.4	50	-
4/20, 22, 5/4	>140.0	1.6-2.0	>2.44	9.5	<68	-
9/21, 22	128.2	1.7	2.30	6.6	52	-
9/28, 29	85.2	1.5-1.6	2.13	4.7	56	-
12/16, 17	47.6	1.5-2.0	1.44	5.7	120	13.75
12/23, 29	66.2	1.5-1.8	1.49	4.7	71	6.62
1988						
2/22, 25	83.3	1.5-1.6	2.39	5.7	68	8.44
3/21, 23	83.3	1.3	2.07	7.6	91	6.21
4/14, 15	88.9	1.3	2.53	5.7	64	7.11
5/10, 11	76.7	1.3	1.92	3.8	49.5	6.95
5/25, 26	79.5	1.3	2.83	5.7	72	7.42
6/2	55.6	1.4-1.7	2.17	3.8	68	9.29
6/6, 9	68.1	1.6	3.32	5.7	84	7.62
6/20	53.0	1.3	1.56	3.8	72	8.18
7/6, 12, 14	125.7	1.7	2.94	7.6	60	3.90
8/9, 10	88.6	1.8	2.54	5.7	64	5.51
8/30, 31	88.9	1.6	2.30	5.7	64	6.16
9/19, 20	-	1.5-1.8	3.26	5.7	-	-
10/3, 20	102.2	1.7	3.97	3.8	37	7.77
10/26, 27	74.9	1.8	2.59	5.7	76	8.64
11/1, 2, 3	115.4	1.8-2.0	2.99	5.7	49	6.64
11/14, 15	98.1	1.5-1.8	3.56	5.7	58	7.26
11/28, 12/1, 5	102.6	1.5-1.8	3.77	5.7	56	8.73
12/14, 5	73.4	1.7-2.8	3.65	5.7	78	13.08

from 1.3 to 2.7 (Table 62).

Sulfuric Acid Addition. The amount of acid used to attain the target pH ranged from 1.44 Mg to 4.3 Mg (i.e., 1.44 x 10^6 to 4.3 x 10^6 g or 1.44 to 4.3 metric tonnes) to produce a volume of recovered coagulant ranging from 50 to 264 m^3 (13,000 to 70,000 gal). Normalization of acid doses to the mass of the recovered iron (i.e., kg H_2SO_4/kg Fe recovered) was performed where the iron concentration of recovered coagulant was available and is presented in Table 62. Normalized acid doses ranged from 3.90 to 13.75 kg H_2SO_4/kg recovered Fe, averaging 7.91 kg H_2SO_4/kg of Fe recovered (±2.39 kg H_2SO_4/kg of Fe) for a total of 18 batches. Based on dissolution of iron in the form of $Fe(OH)_3 \cdot 3H_2O$ to Fe, stoichiometry would predict an acid dose of 1.31 kg/kg of Fe. Therefore, the normalized acid dose typically used at the facility is 3- to 10-fold higher than predicted by stoichiometry. Comparison to other coagulant recovery data reported earlier indicates that these values are, however, typical of those expected in coagulant recovery operations. Other acid-demanding constituents in the sludge, such as heavy metal oxides and hydroxides, can account for a fraction of this acid demand. The excess acid is added to compensate for the low detention time in the acid contact basin, but may result in excess free acid in the recovered product (i.e., high acidity). Further investigation of acid application rates, acid-contact basin detention time and fractional iron recovery may be necessary to optimize the coagulant recovery process. However, given the lack of control over the suspended solids concentration of the sludge, excess acid may be required to recover the maximum amount of iron from the sludges.

Polymer Addition. Acidified sludge solids are conditioned with a nonionic polymer prior to dewatering. Polymer application rates were highly variable, ranging from 37 to 150 ppm (volumetric basis), as presented in Table 62. In September 1988, a change was made to another nonionic polymer which was used for production of one batch of recovered coagulant. During the initial period of use of this recovered coagulant, the dual-media filters experienced significant increases in head loss which were attributed to the change in polymer. The batch of recovered coagulant was subsequently wasted from the recovered coagulant storage basin. Although this phenomena was attributed to the change in polymer in the recovery process, no definitive explanation for

the impact was determined. However, carryover of the polymer to the water treatment system may have impacted the coagulation process, increasing the solids load to the filters. Consequently, the original nonionic polymer is currently being employed to aid dewatering of acidified sludge solids in the recovery process.

Recovered Coagulant Quality

As discussed previously, the coagulant recovery process is controlled by the pH value in the effluent of the acid-contact basin. The pH value of the recovered coagulant solution is also measured following storage to document any changes with time and is indicative of the pH value of the recovered coagulant solution at the time of application at the rapid mix unit. As presented in Table 63, this 24-h pH value ranged from 1.2 to 2.5, averaging 1.6 for all 29 runs, while the average 24-h pH value (arithmetic average of pH values) during 1988 was 1.8. The pH value in the effluent of the acid-contact basin was similar to the 24-h pH value, indicating little change in the recovered solution pH as a function of time.

Manganese concentrations in the recovered coagulants were determined for nearly all batches, while iron concentrations were determined for all batches of recovered coagulant since December 1987. It should be noted that these concentrations were determined on grab samples collected randomly. As presented in Table 63, iron concentrations in the recovered coagulant ranged from 2200 to 6400 mg/L as Fe and are typical of concentrations found in other investigations of iron recovery (Figure 41). These iron concentrations were used to determine mass application rates for the recovered coagulant. Compared to commercial ferric sulfate with an iron concentration of about 160,000 mg/L, recovered coagulants were 25- to 73-fold lower in concentration than the commercial ferric sulfate product.

Manganese concentrations presented in Table 63 ranged from 50 to 500 mg/L in recovered coagulants. These concentrations were employed to investigate projected incremental increases in manganese concentrations at the point of coagulant addition attributable to use of recovered coagulants. Other possible contaminants in the recovered products, such as heavy metals and

Summary of Recovered Coagulant pH, Manganese and Iron Concentration Reported in Full-Scale Operation at Athens, TN

Date		pH	Mn, mg/L	Fe, mg/L	Normalized Mn mg/kg Fe
1985	10/7, 8	1.3	60	-	-
	11/1, 5, 12	2.0	120	-	-
	12/4, 5, 17, 30	1.5-1.75	83	-	-
1986	10/30, 11/10	1.9	-	-	-
1987	1/6, 7, 14	1.9	-	-	-
	3/4, 5, 25	-	125	-	-
	4/20, 22, 5/4	-	175	-	-
	9/21, 22	1.9	90	-	-
	9/28, 29	1.7	-	-	-
	12/16, 17	2.0	400	2200	181818
	12/23, 29	2.15	200	3400	58823
1988	2/22, 25	1.8	150	3400	44118
	3/21, 23	1.8	180	4000	45000
	4/14, 15	1.2	120	4000	30000
	5/10, 11	1.4	120	3600	30000
	5/25, 26	1.6	110	4800	22917
	6/2	1.6	120	4200	28571
	6/6, 9	1.6	60	6400	9375
	6/20	1.75	120	3600	33333
	7/6, 12, 14	1.4	120-140	5600-6400	18750-25000
	8/9, 10	1.7	200	5200	38462
	8/30, 31	2.0	50	4200	11905
	9/19, 20	1.7	200	4200	47619
	10/3, 20	1.4	200	4800-7200	41667-38462
	10/26, 27	1.9	200	4000	50000
	11/1, 2, 3	1.9	100	3900	25641
	11/14, 15	2.1	500	5000	100000
	11/28, 12/1, 5	1.9	200	3800	52632
	12/14, 15	2.5	200	3800	52637

organics, have not been investigated directly.

Impacts of Recovered Coagulants on Daily Plant Operations and Performance

Plant operations and performance data were compared during periods of commercial and recovered coagulant use. Use of recovered coagulants can impact both chemical addition rates and finished water quality, due to, for example, higher coagulant acidity and contaminant concentrations, as compared to commercial coagulant. Furthermore, the lower iron concentration in the recovered coagulant may impact its effectiveness. Increased lime for pH and alkalinity adjustment may be required due to the high acidity of the recovered product, while increased chlorination may be needed to oxidize higher levels of manganese and ferrous iron that may be present. Coagulant, lime and chlorine application rates during periods of recovered and commercial coagulant use were compared, as well as finished-water iron and manganese concentrations.

The performance of the sedimentation and rapid sand filtration system were addressed to determine possible impacts associated with use of the recovered product. Using raw, settled and filtered water turbidity, conclusions were drawn as to the efficiency of the rapid mix, sedimentation and filtration processes by comparison of performance during use of recovered and commercial coagulant products. Furthermore, the length of rapid sand filtration runs were examined to determine impacts on the rate of head loss and turbidity breakthrough.

The period of January through December 1988 was examined in detail since the most intensive coagulant recovery was practiced over this period and, as indicated previously, the current system employed was put on line in December 1987. Since January 1988, 19 coagulant recovery runs have been conducted at the site.

The total flow treated with each coagulant on a monthly basis in 1988 is presented in Table 64, along with the total for the year. An average of about 2.38×10^5 m^3/mo (63×10^6 gal/mo) were treated, totaling 2.87×10^6 m^3 (757.4×10^6 gal) for 1988. The monthly percentage of flow treated with recovered

TABLE 64

Total Flow Treated Using Commercial and Recovered Coagulants at Athens, TN Plant in 1988

Month	Flow Treated, m^3/mo		Flow Treated, 10^6 gal/mo		Percent Total Flow Treated	
	Commercial Coagulant	Recovered Coagulant	Commercial Coagulant	Recovered Coagulant	Commercial Coagulant	Recovered Coagulant
January	11,250	3,933	42.582	14.888	74.1	25.9
February	14,691	740	55.605	2.8	95.2	4.8
March	8,618	9,388	32.618	35.535	47.9	52.1
April	8,841	5,509	33.465	20.851	61.6	38.4
May	10,169	5,795	38.488	21.934	63.7	36.3
June	7,524	10,272	28.477	38.878	42.3	57.7
July	9,821	6,508	37.173	24.634	60.1	39.9
August	12,890	3,885	48.789	14.705	76.8	23.2
September	14,620	3,148	55.336	11.916	82.3	17.7
October	9,345	6,603	35.37	24.991	58.6	41.4
November	9,401	6,518	35.581	24.671	59.1	40.9
December	12,932	7,709	48.948	29.18	62.7	37.3
Total	130,101	70,009	492.432	264.983	-	-
Average	10,842	5,834	41,036	22,082	65.0	35.0

coagulant ranged from about 5 percent in February to 39 percent in June, with 35 percent of the total flow in 1988 being treated with recovered coagulant. A total of 1.86 x 10^6 m^3 (492 x 10^6 gal) of raw water was treated with commercial coagulant in 1988, while 1.00 x 10^6 m^3 (265 x 10^6 gal) was treated with recovered coagulant. This high frequency of coagulant recovery and use in 1988 allowed for detailed investigation of chemical application rates and treatment plant performance during periods of commercial and recovered coagulant use.

Chemical Addition

Coagulant Application. Optimum coagulant doses were determined on a daily basis using standard jar test procedures for both commercial and recovered coagulants. Commercial coagulant application rates were reported herein using gravimetric concentrations, i.e., as mg/L as Fe. Since the iron concentration of the recovered coagulant was not always known, the volumetric application rate calculated by plant personnel has been reported. Therefore, comparison of mass application rates for commercial and recovered coagulants was not always possible, but was performed where applicable iron concentrations were determined.

As presented in Figure 57, typical commercial coagulant doses ranged from 4 to 10 mg/L as Fe, with perturbations to as high as 31 mg/L as Fe during periods of poor raw water quality. To examine the predominant mechanisms governing coagulation at the plant, coagulant dose and pH data were compared to predominance areas on an iron coagulation diagram, as presented in Figure 4. In the pH range of 7.5 to 8.0 and a coagulant dose of about 4 to 10 mg/L as Fe (i.e., Fe = 7.2×10^{-5} to 1.8×10^{-4} M and log[Fe] = -4.14 to -3.75), sweep coagulation is the predominant coagulation mechanism. That is, coagulant is added at doses sufficiently high to cause a rapid precipitation of iron hydroxide solids which act to agglomerate and remove particulate and soluble contaminants from the raw water.

Recovered coagulant doses through 1988, as presented in Figure 58, ranged from about 1.0 to 2.4 mL/L. Large perturbations in recovered coagulant doses were not observed, as compared to those observed in the commercial coagulant doses

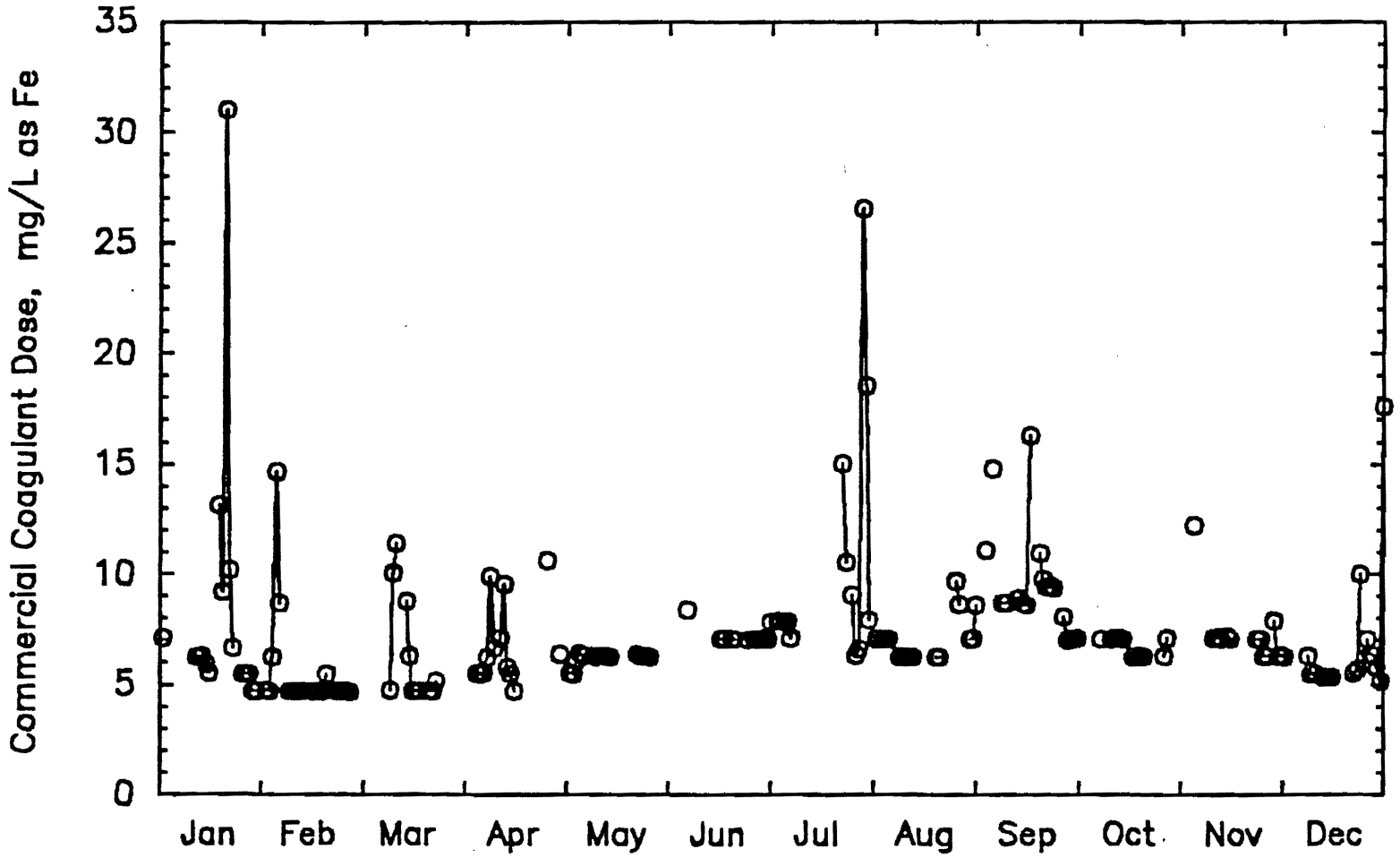

FIGURE 57. 1988 Commercial Coagulant Dose (mg/L as Fe) at the Athens, TN Water Treatment Plant

for two reasons. First, recovered coagulant use was apparently limited to, or only used by chance, during periods when raw water quality was typical of annual averages, with few exceptions. Secondly, when raw water quality deteriorated during use of recovered coagulant, commercial coagulant was added simultaneously to ensure maintenance of finished water quality. During the period 1988, however, this occurred on only 9 occasions totalling 20 days of operation.

Using the iron concentrations reported for recovered coagulants, the volumetric application rates presented in Figure 58 were converted to mass application rates for comparison of commercial and recovered coagulant doses, as presented in Figures 59 and 60 for the periods of January-June and July-December 1988, respectively. The application rates for commercial and recovered coagulants were essentially the same, with a few periods where the recovered coagulant dose was lower than the commercial coagulant dose during periods of consistent raw water quality. The lower recovered coagulant doses reported for the end of February, the end of March and mid-April were during periods when recovered and commercial coagulants were used simultaneously. In general, coagulant doses ranged from about 4 to 10 mg/L as Fe for each coagulant, with several periods of dramatic fluctuations due to variations in raw water quality. Based on comparison of the mass application rates of the coagulants alone, commercial and recovered coagulants appeared to be equally effective. However, chlorine and lime application rates, as well as finished water quality and plant operations must be investigated during these periods of use to adequately determine the relative effectiveness of recovered coagulants as compared to hydrated commercial coagulants.

Lime Application. Typical hydrated lime [$Ca(OH)_2$] doses ranged from about 15 to 40 mg/L with extreme fluctuations during periods of poor raw water quality to maintain a final pH ranging from about 7.6 to 8.2, similar to the raw water pH value of 7.5 to 8.2. Lime addition data over the period of January through December 1988 are presented in Figure 61, along with the indicated periods of recovered coagulant use. The data indicate the high degree of scatter inherent to the addition of lime due to changes in raw water quality and coagulant application rates.

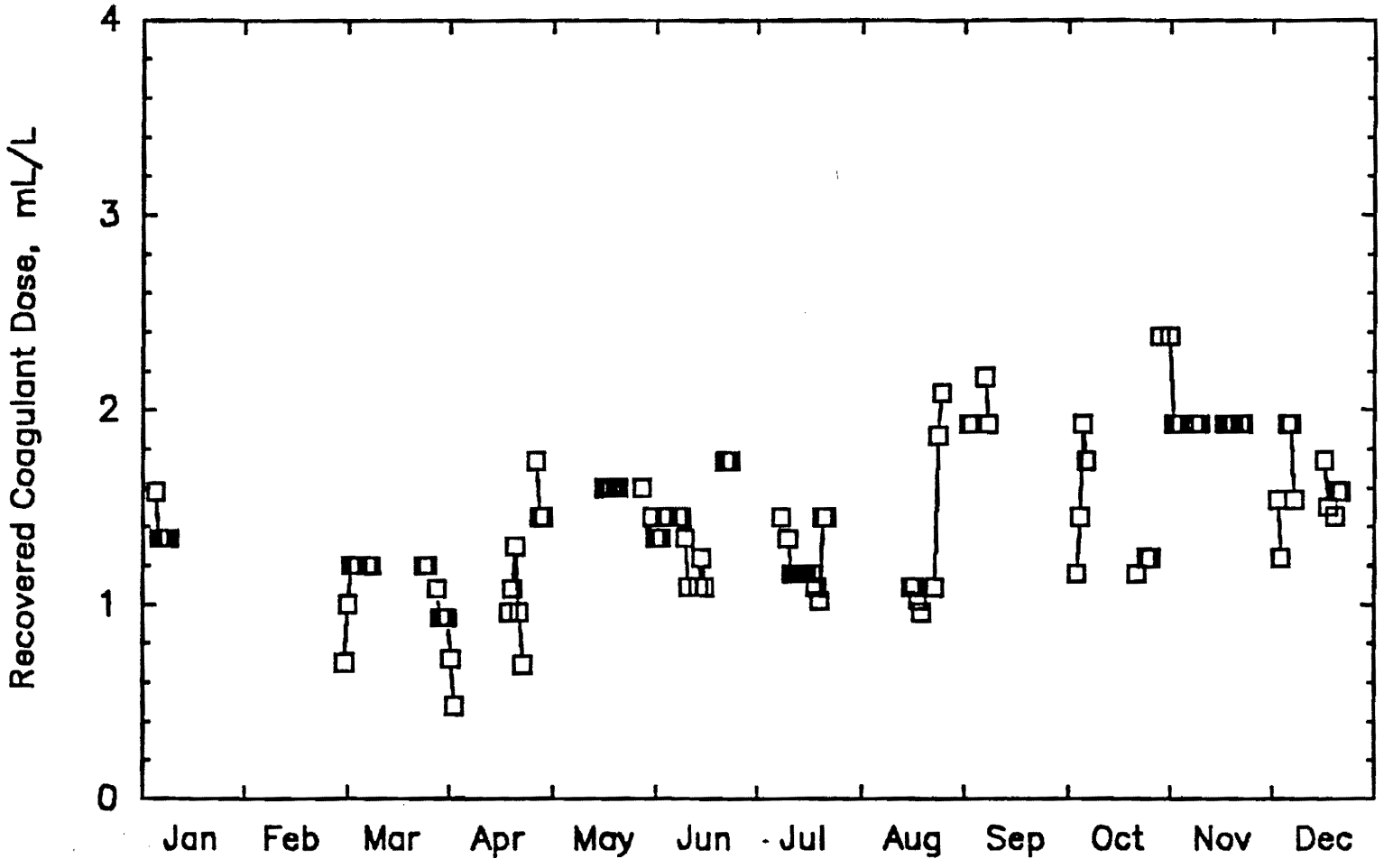

FIGURE 58. 1988 Recovered Coagulant Volumetric Application Rate (mL/L) at the Athens, TN Water Treatment Plant

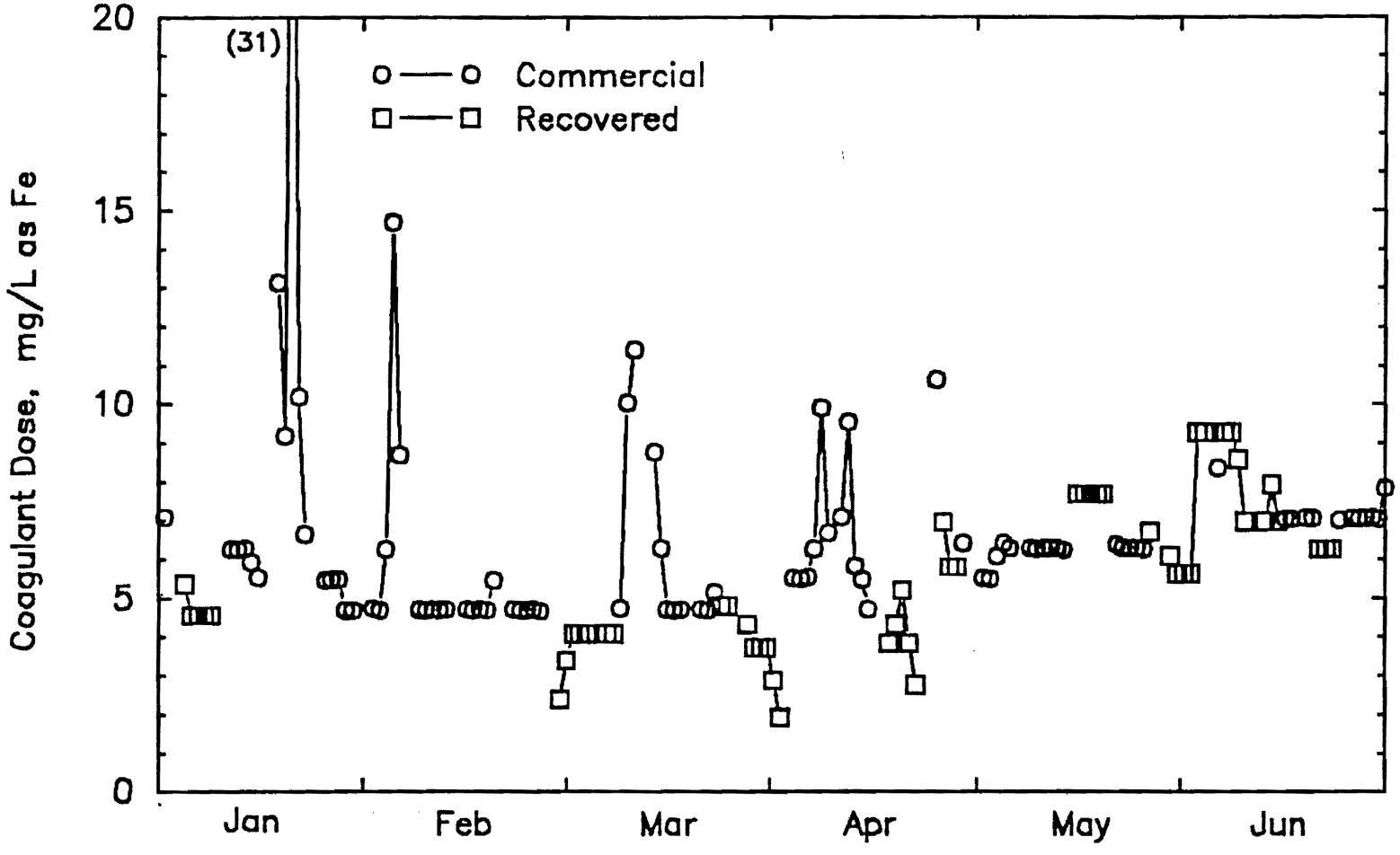

FIGURE 59. Commercial and Recovered Coagulant Mass Application Rates for January through June of 1988

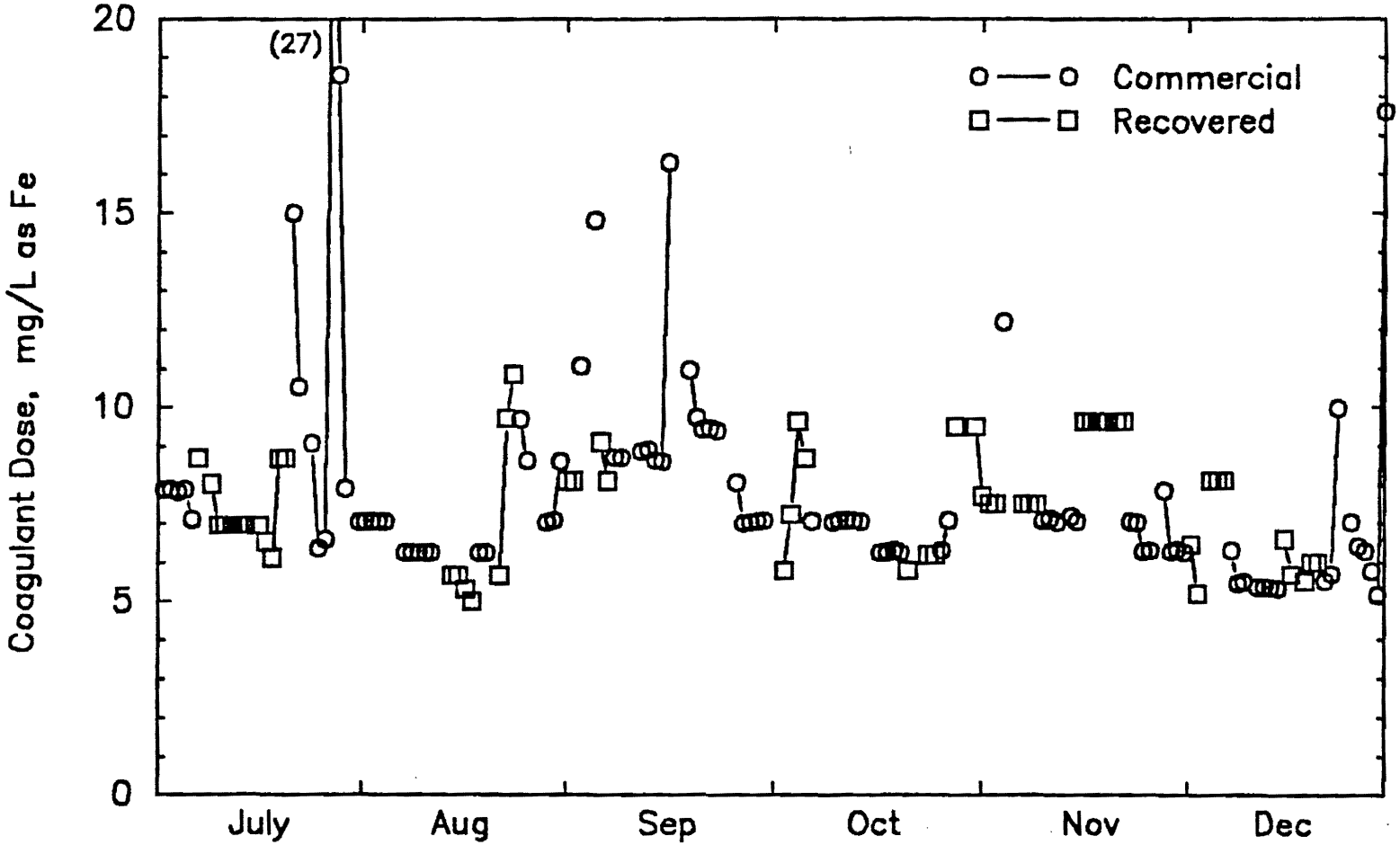

FIGURE 60. Commercial and Reocvered Coagulant Mass Application Rates for July through December of 1988

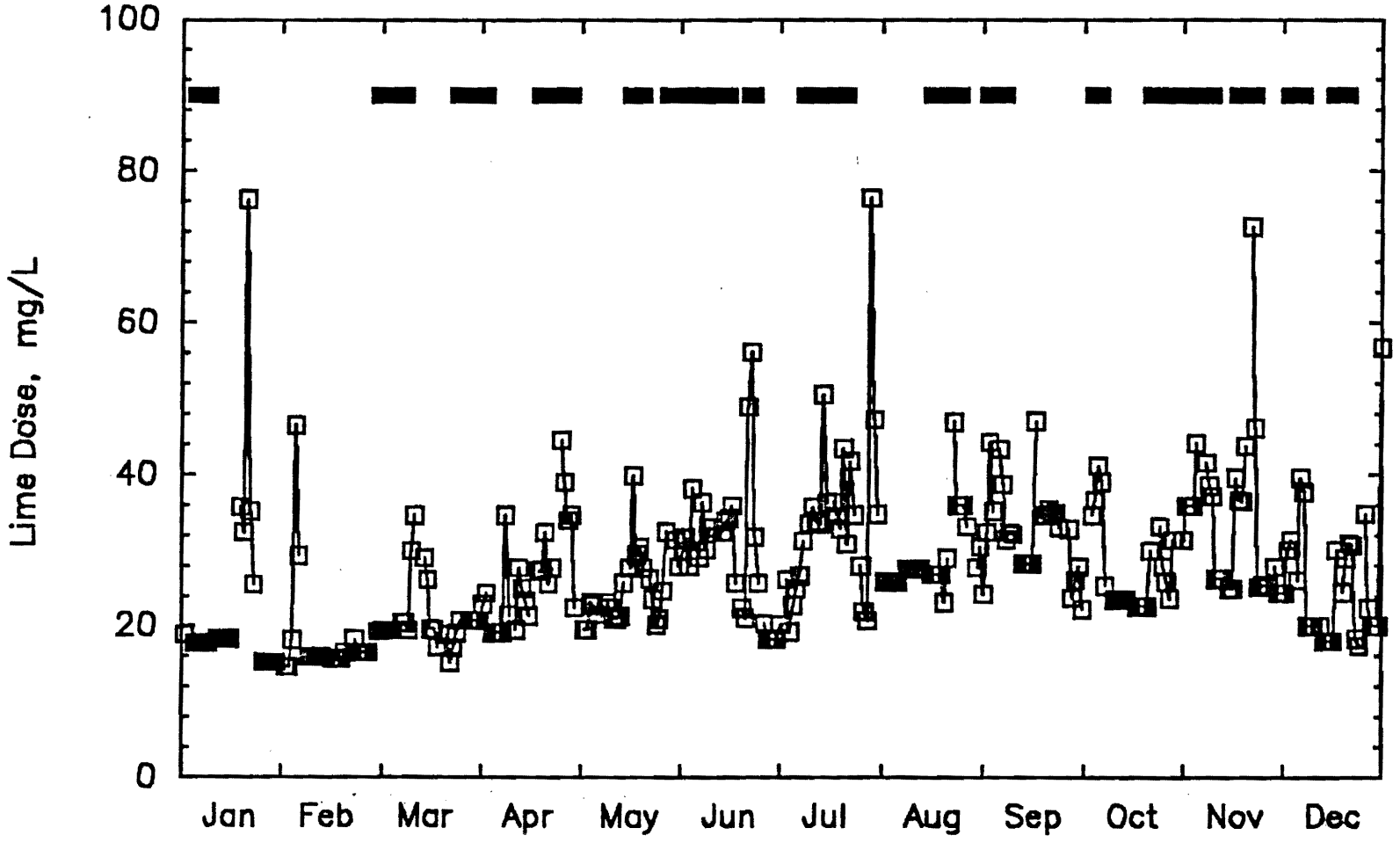

FIGURE 61. 1988 Lime Dose and Indicated Periods of Recovered Coagulant Use

Inspection of lime application rates during periods of commercial and recovered coagulant use indicated that an increased lime dose may occur during use of recovered coagulants. For example, during the period of May through June 1988, the lime dose appeared to increase during use of recovered coagulant and decreased when use of commercial coagulant resumed. To investigate this point further, the average lime doses during use of commercial and recovered coagulants were investigated on a monthly basis for 1988.

Monthly average application rates are presented in Figure 62 and indicate the impact of recovered coagulant use on the rate of lime application. For example, lime application rates in May 1988 were about 13 mg/L and 21 mg/L during use of commercial and recovered coagulants, respectively. With the exception of January, lime application during recovered coagulant use is equal to or higher than that during use of commercial coagulant. In January, commercial coagulant was used to treat 74.1 percent of the water, and a severe deterioration of the raw water was experienced, thereby increasing the average lime application rates during use of the commercial coagulant.

The average annual lime dose during use of commercial coagulant was calculated to be 23.2 mg/L, compared to 27.6 mg/L during use of recovered coagulant. This corresponds to an 18.7 percent increase in lime application rate during use of recovered coagulant for 1988. Although raw water quality was not integrated into the above analysis, a similar analysis was conducted excluding data when raw water turbidity exceeded 20 NTU. Results of this analysis were similar to those obtained using all of the data, indicating that raw water quality can not explain the increased demand, or the apparent scatter, in the data.

From the above analysis, it was concluded that use of recovered coagulant caused an increase in the lime dose for pH and alkalinity adjustment. Quantification of the increased demand is more difficult. However, using the average annual lime doses for commercial and recovered coagulants in 1988, an 18.7 percent increase was predicted during use of recovered coagulants.

The increase in lime dose during recovered coagulant use can be attributed to

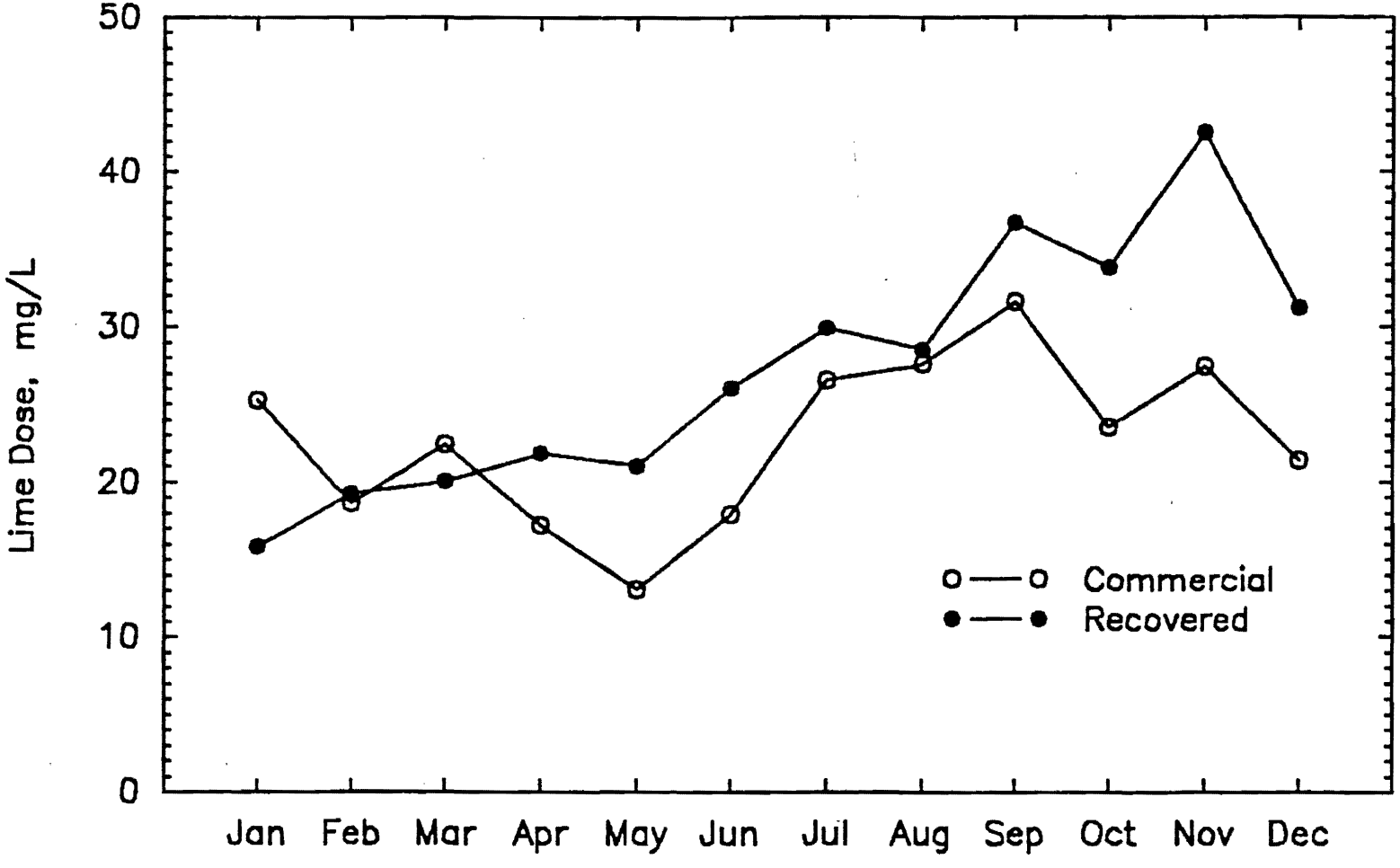

FIGURE 62. 1988 Monthly Averaged Lime Doses During Use of Commercial and Recovered Coagulants

a higher free acidity of the recovered coagulant product over that of the commercial coagulant. As indicated by acid application rates discussed previously, excess acid is added in the recovery process at 3 to 10 times the stoichiometric rate, part of which may exist as free acidity in the recovered product. Since the mass application of iron was similar for commercial and recovered coagulants (see Figures 59 and 60), the alkalinity consumed in the coagulation process by the precipitation of $Fe(OH)_3 \cdot 3H_2O$ would be equivalent for each coagulant. Therefore, the increased lime dose must be attributable to a higher free acidity in the recovered coagulant, as compared to the commercial product.

Chlorine Application. Pre-chlorination is provided for disinfection and oxidation of manganese and ferrous iron in the raw water, while post-chlorination is provided for final disinfection and to maintain a residual in the distribution system. Increased levels of manganese or ferrous iron in the raw water or from addition of the recovered coagulant can have a significant impact on required chlorine dose to maintain a residual throughout the treatment process. As presented in Figure 63, total chlorine additions typically range from 3 to 6 mg/L, with higher application rates during periods of poor raw water quality. The operational strategy is to maintain a residual chlorine level in the plant effluent ranging from 2.0 mg/L as Cl_2 to 2.8 mg/L as Cl_2 and about 1.0 mg/L as Cl_2 in the distribution system.

Chlorine application rates show the same trends as lime addition, following raw water quality and coagulant addition, and also exhibiting a high degree of scatter. For this reason, a definitive correlation between recovered coagulant use and chlorine demand could not be established. Therefore, chlorine application rates were investigated in the same manner as lime, i.e., by determining average monthly application rates for pre- and post-chlorine addition during use of each coagulant.

Monthly averaged pre-chlorine doses, presented in Figure 64, ranged from about 1.7 to 3.2 mg/L as Cl_2 and were higher during use of recovered coagulant for January, February and September through December 1988. During the period of April through August, the doses were essentially equal. An annual average dose of 1.99 mg/L was calculated during commercial coagulant use, compared to

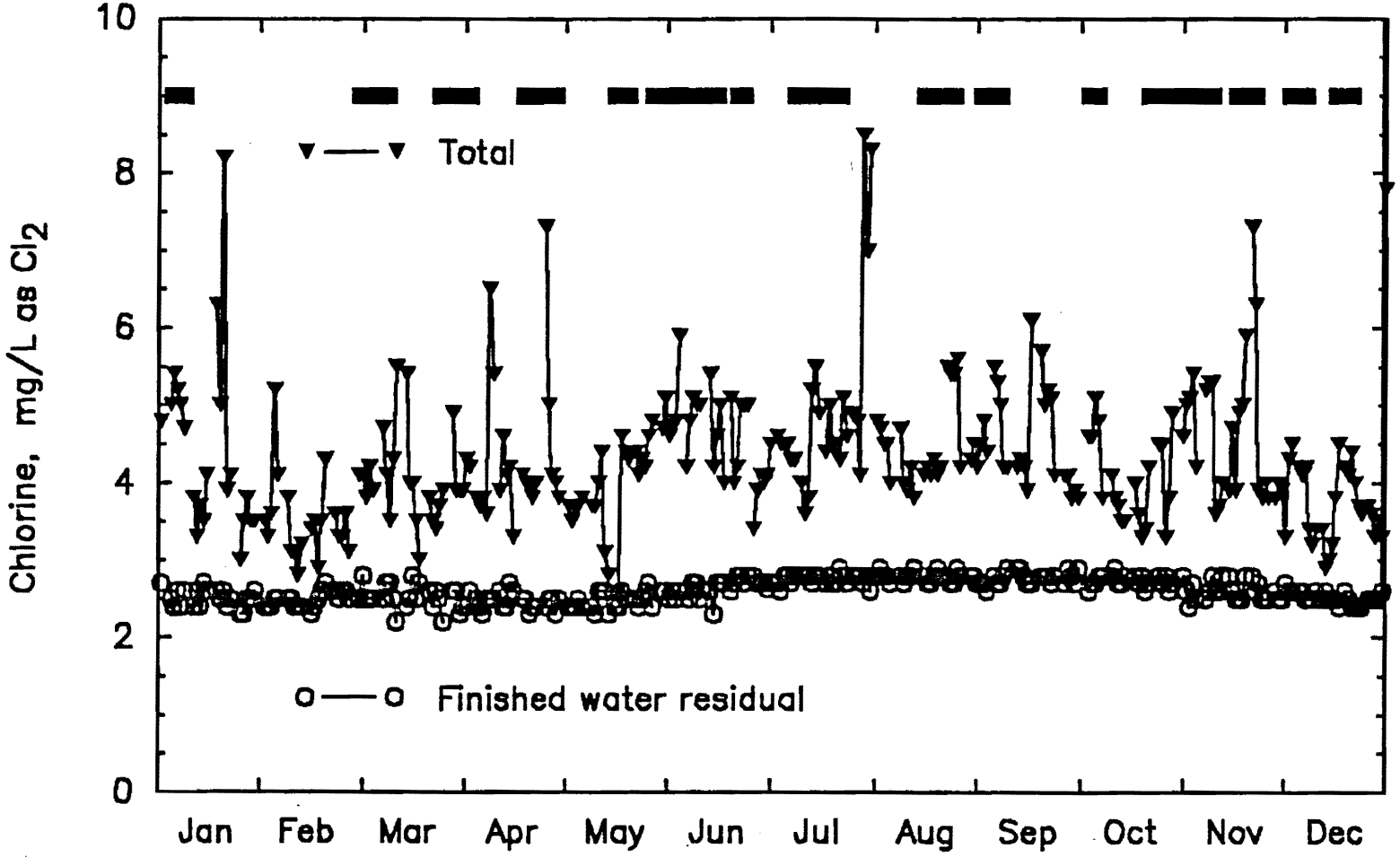

FIGURE 63. Total Chlorine Dose, Finished Water Residual Chlorine Concentration and Indicated Periods of Recovered Coagulant Use During 1988

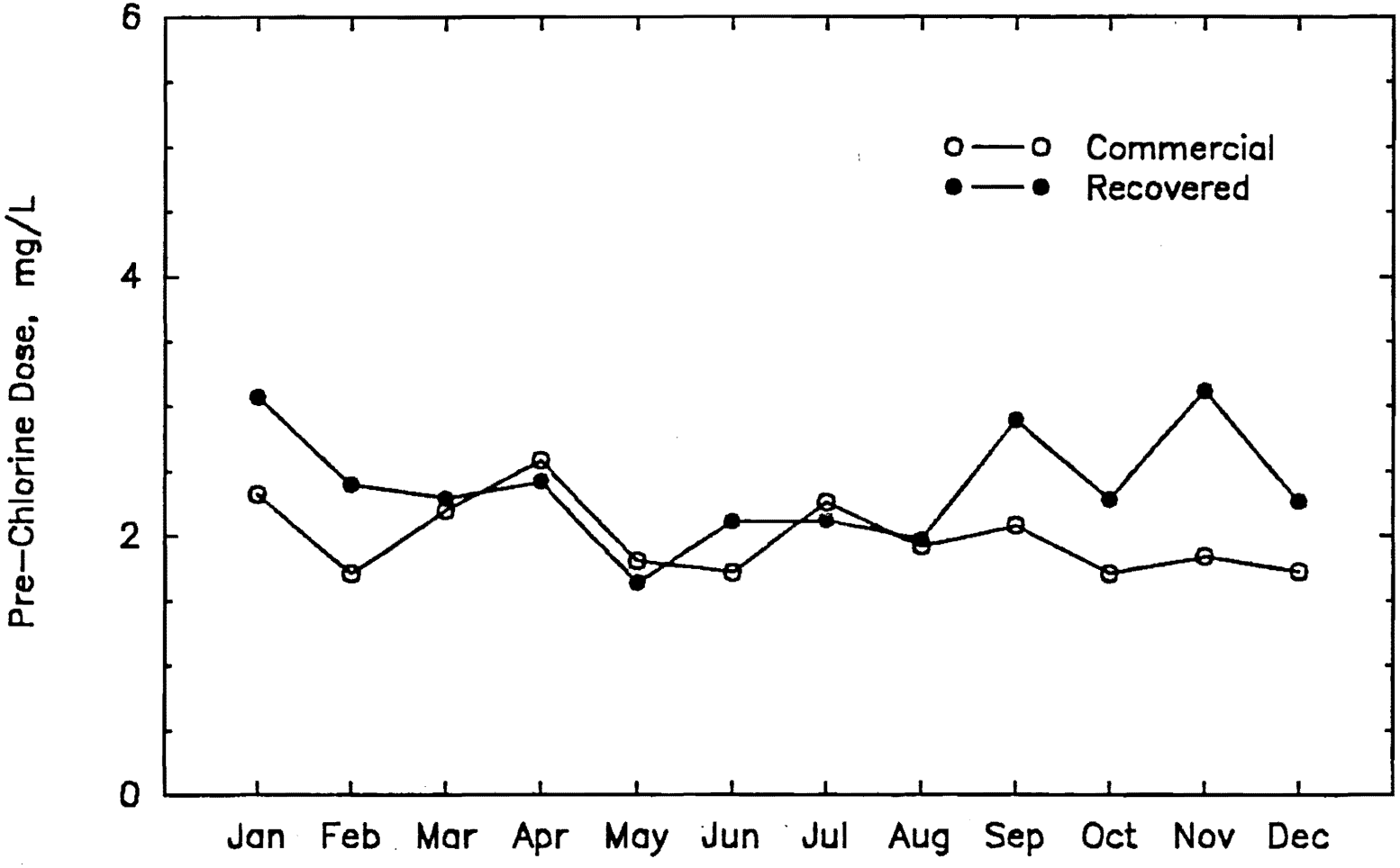

FIGURE 64. 1988 Monthly Averaged Pre-chlorine Doses During Use of Commercial and Recovered Coagulants

2.31 mg/L during use of recovered coagulant. This corresponds to a 16.1 percent increase in the average pre-chlorine dose attributable to the use of recovered coagulant.

Post-chlorination was also examined, with monthly averages presented in Figure 65. Comparison during periods of recovered and commercial coagulant use indicated that post-chlorination was not impacted as dramatically as pre-chlorination. The average monthly dose ranged from 1.6 to 2.9 mg/L as Cl_2 and was similar during commercial and recovered coagulant use. Examination of annual average doses indicated an 8.7 percent increase in required post-chlorination doses during use of recovered coagulant; i.e., 2.07 mg/L as Cl_2 using commercial coagulant versus 2.25 mg/L as Cl_2 during use of recovered coagulant. Therefore, the total chlorine dose required during use of recovered coagulant averaged about 0.50 mg/L higher than during commercial coagulant use, a 12.3 percent increase attributable to use of the recovered coagulant.

As discussed previously, manganese concentrations in the recovered coagulant ranged from about 50 to 500 mg/L, and averaged 169 mg/L during 1988. Using the maximum reported manganese concentration of 500 mg/L and a recovered coagulant dose of 1.5 mL/L, a projected incremental increase of manganese in the water at the point of coagulant addition attributable to the use of the recovered coagulant can be calculated to be 0.75 mg/L. Since raw water manganese concentrations average about 0.1 mg/L, use of recovered coagulant would constitute nearly an eight-fold increase in manganese concentration at the point of coagulant addition. An increased chlorine dose of about 1.0 mg/L as Cl_2 of would be predicted for oxidation of the 0.75 mg/L of manganese projected from use of the recovered coagulant. Using the average manganese concentration (i.e., 169 mg/L) and an application rate of 1.5 mL/L, about 0.25 mg/L of manganese would require 0.34 mg/L of Cl_2 for oxidation. The above analysis indicated that a large portion of the increased chlorine demand experienced at the plant during use of recovered coagulants may be due to oxidation of manganese.

Contamination of the recovered coagulants by reduced compounds, such as Mn^{++} and Fe^{++} is responsible for a majority of the impact on chlorine application.

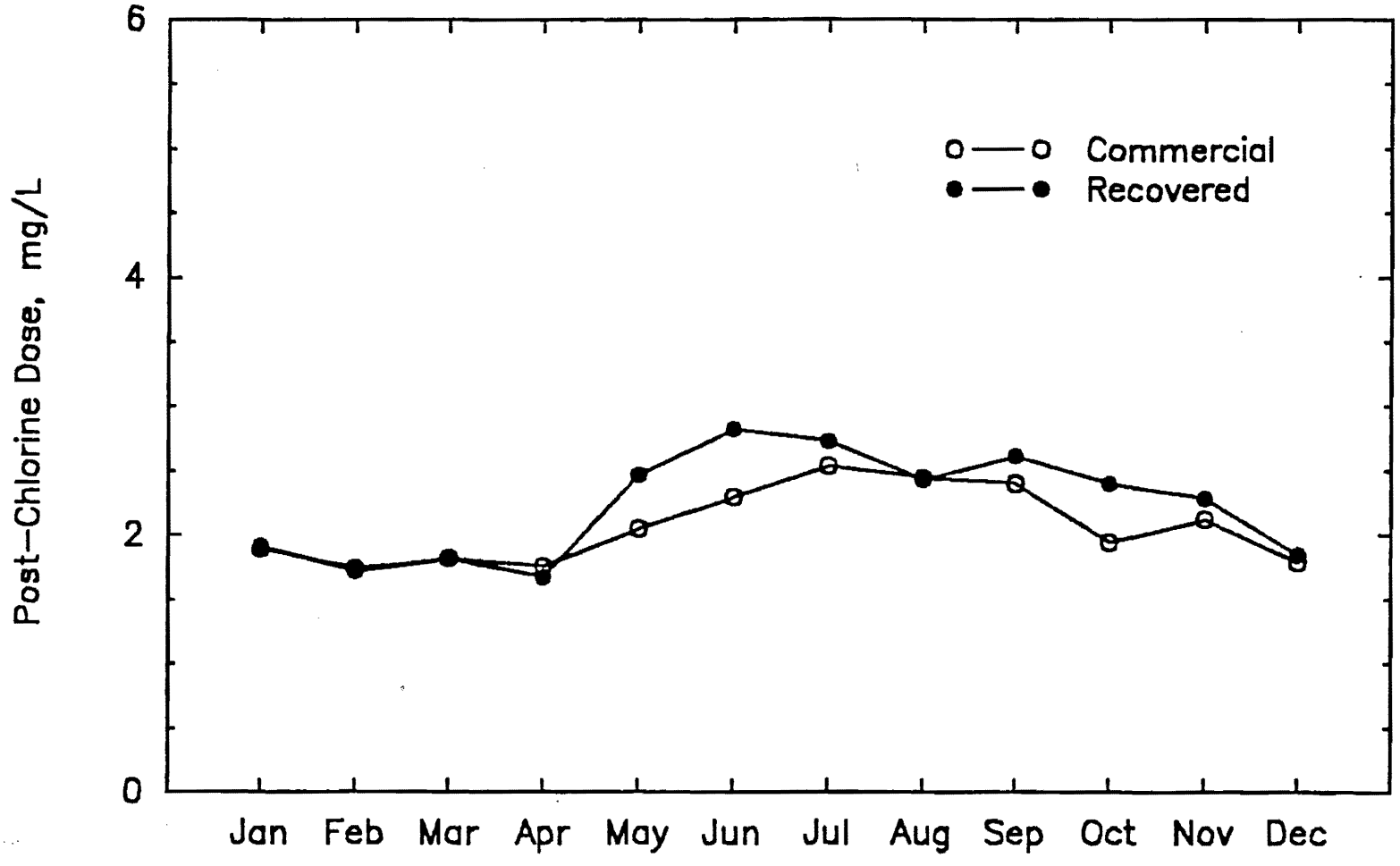

FIGURE 65. 1988 Monthly Averaged Post-chlorine Dose During Use of Commercial and Recovered Coagulants

The high pH maintained in the treatment plant (i.e., 7.5 to 8.0) favors the oxidation of these reduced compounds, thereby decreasing the impact of increased concentrations of reduced compounds in the recovered coagulant or raw water (Stumm and Morgan 1981).

Impact of Recovered Coagulant Use on Sedimentation and Filtration Processes

The turbidity of settled and finished waters was utilized to investigate the impact of the use of recovered coagulant on the sedimentation and filtration processes. Recovered coagulant may impact the destabilization and agglomeration of raw water particles and floc size and density may also change with use of the recovered coagulant due to differences in the chemical forms of commercial and recovered coagulants. Although such changes can not be documented directly, they may have considerable impact on the efficiency of the sedimentation and filtration processes for the removal of particulate matter and were examined indirectly using turbidity values as an indicator parameter.

Sedimentation Process. Settled water turbidity is presented in Figure 66 along with the indicated periods of recovered coagulant use. Close investigation of data presented in this figure indicated that there may be an increase in settled water turbidity during use of recovered coagulants. For example, when recovered coagulant was used in the first two weeks of March, settled water turbidity exhibited a substantial increase from about 0.5 NTU to nearly 1.0 NTU. The same trend is observed near the end of October and beginning of November. As recovered coagulant use was initiated, an increase in settled water turbidity was observed until commercial coagulant use resumed. The severe fluctuations in settled water turbidity which occur naturally, however, complicated data analysis and made it difficult to distinguish these differences.

To investigate this point further, the average settled water turbidity was calculated on a monthly basis for periods of commercial and recovered coagulant use, as presented in Figure 67. No consistency in settled water turbidity during periods of commercial or recovered coagulant use was observed. Again, due to extreme variations in settled water turbidity which

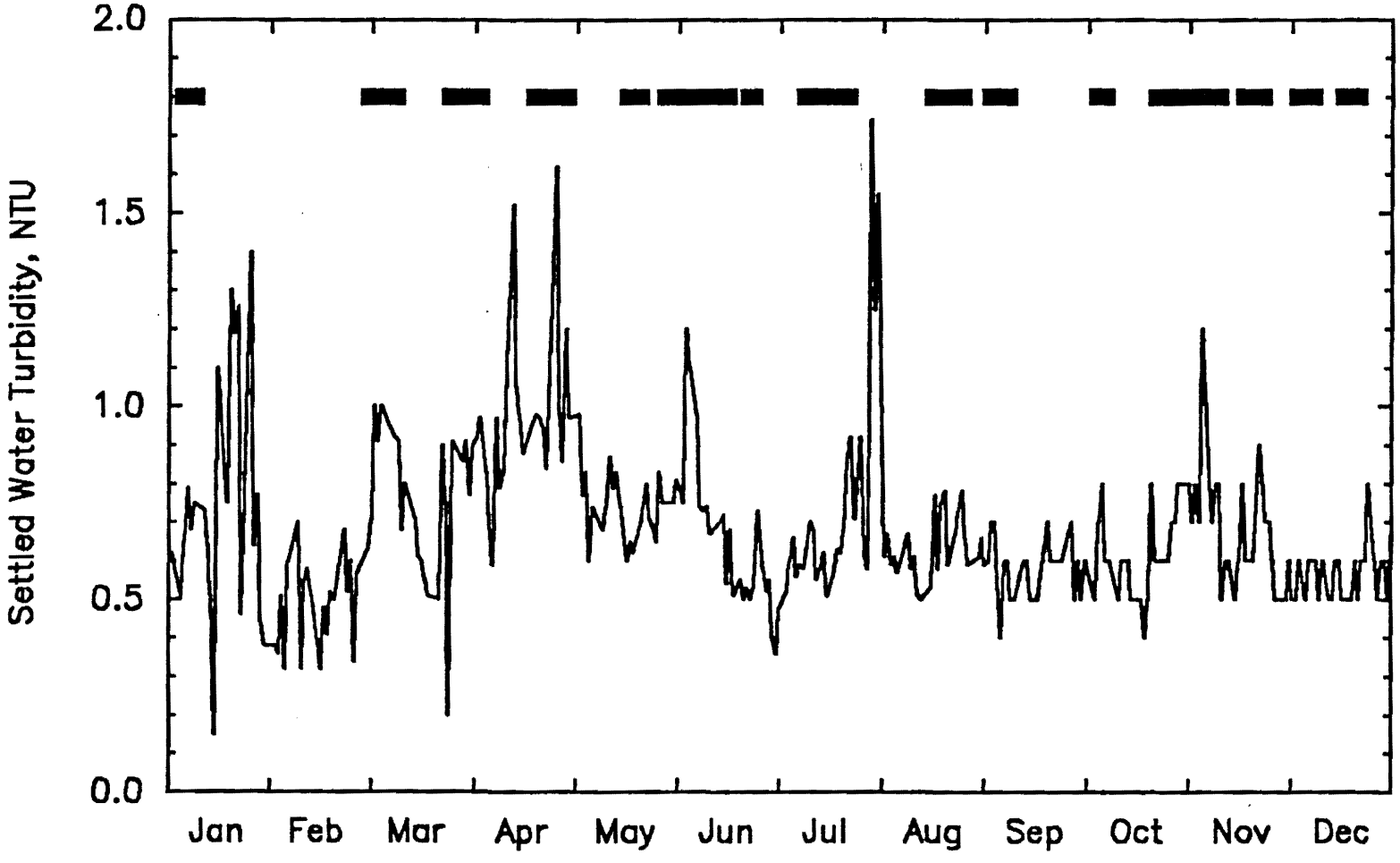

FIGURE 66. 1988 Settled Water Turbidity and Indicated Periods of Recovered Coagulant Use

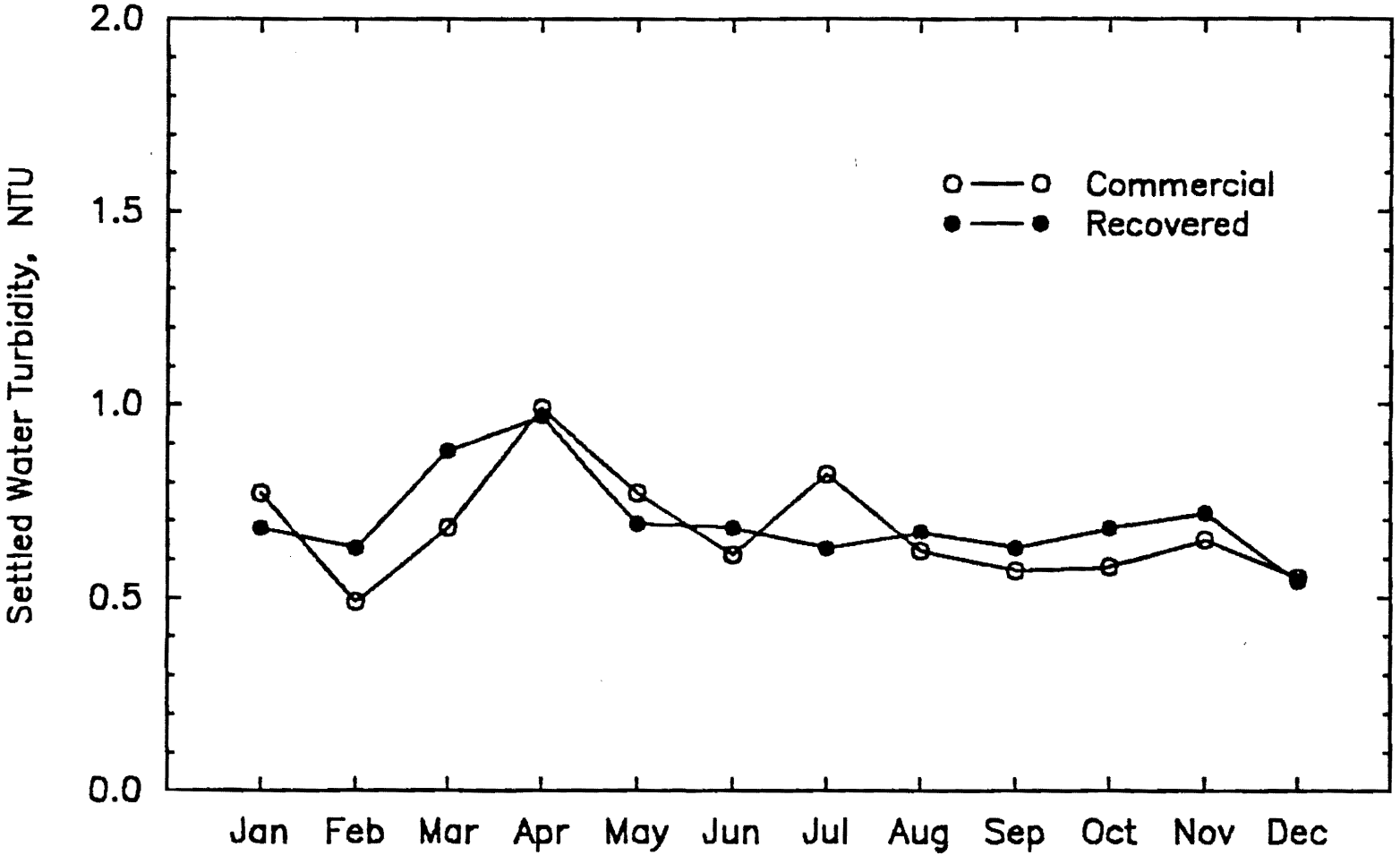

FIGURE 67. 1988 Monthly Averaged Settled Water Turbidity During Use of Commercial and Recovered Coagulants

occur naturally, any impact on settled water turbidity from the use of recovered coagulant was difficult to distinguish. However, from the local areas of increased turbidity exhibited during periods of recovered coagulant use, it was concluded that the efficiency of the sedimentation process deteriorated slightly during use of the recovered product.

Rapid Sand Filtration Operation. Plant personnel indicated that head loss is the controlling factor in the determination of the time a filter will be in operation prior to backwash. Since the filters are backwashed upon reaching a consistent maximum head loss, it was assumed that the length of filter runs can be directly compared during use of commercial and recovered coagulants. The average length of a filter run is determined by the average daily filtration time and the number of days the filter was in operation. The percentage of the filter run time during which recovered coagulant was used was also calculated. For example, 50 percent recovered coagulant use indicates that recovered coagulant was used as primary coagulant for 50 percent of the time that the filter was in operation prior to backwash.

The average lengths of filter runs in 1988 are presented in Figure 68, with the length of filter operation prior to backwash plotted on the date of backwashing. Filter run times ranged from 60 to 120 hours and were dependent on solids loading and raw water temperature. In some instances, filter run time decreased as recovered coagulant use increased. For example, as recovered coagulant use increased in the beginning of March, a sharp decrease in filter run time was observed. Following the return to the use of commercial coagulant, filter run times increased to near the original value of approximately 100 h. This same phenomena is observed to some degree at four other occasions throughout the year, while other periods, such as the end of May through June, do not indicate such an impact from the use of recovered coagulant.

From the data presented above, use of the recovered coagulant has some degree of detrimental impact on the maximum length of a filter run prior to backwash, as indicated by headloss across the filter. However, quantification of this impact and the mechanism of the impact can not be determined from the above analysis. To further investigate this point, the impact of settled water

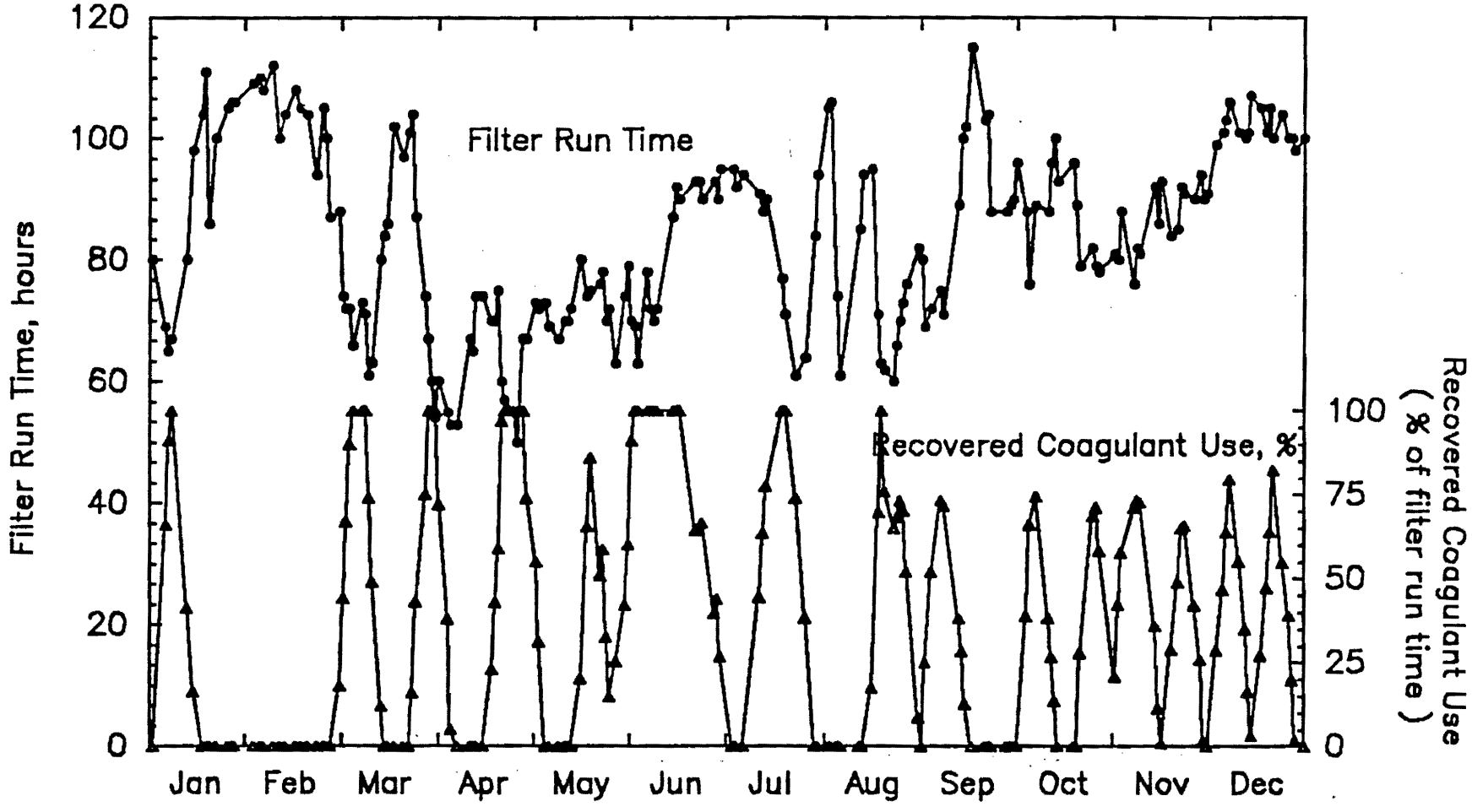

FIGURE 68. Length of Rapid Sand Filtration Runs Prior to Backwash and the Percentage of This Time That Recovered Coagulants Were in Use During 1988

turbidity and raw water temperature fluctuations on the length of a filter run were investigated and compared to periods of use of the recovered coagulant.

Raw water temperature, presented earlier in Figure 56, is recorded daily and can be related to water viscosity through use of a standard table. Weber (1972) showed that head loss across the filter is directly proportional to viscosity. The head loss and, therefore, the length of a filter run can be affected dramatically by seasonal changes in raw water temperature. For example, the 14°C seasonal change in temperature from 8°C to 22°C observed in 1988 would correspond to a 31 percent decrease in viscosity from 1.386 to 0.955 cp, with a proportional change in head loss. From the above example, the impact temperature can have on the length of a filtration run can be considerable, but should be seasonal in nature. However, the impact of these seasonal temperature changes appears to be insignificant compared to the impact of settled water turbidity on the length of a filter run.

Settled water turbidity is reported as a daily average and is an indication of the efficiency of the sedimentation process for solids removal and is proportional to the particle load on the filters. An increase in the turbidity to the filters would cause an increase in the rate of accumulation of head loss and a subsequent decrease in the time of a filter run. Due to the length of a filter run and the non-continuous plant operation, a filter is typically in operation for an average of 5 days previous to backwashing. Therefore, several days of increased turbidity just prior to backwashing a filter would cause a substantial increase in solids loading on the filter. Whereas, one day of increased turbidity may have little impact on the filter run time.

The length of a filter run and settled water turbidity are presented in Figure 69, with an arbitrarily-selected tenth-order regression line fitted through each data set. In general, when settled water turbidity increased, the filter run time decreased and vice versa. For example, as the turbidity began to show a general upward trend in mid-February, the length of a filter run consequently experienced a substantial decrease. The same trends are observed throughout the period 1988, indicating that the filter run time is highly dependent on solids loading.

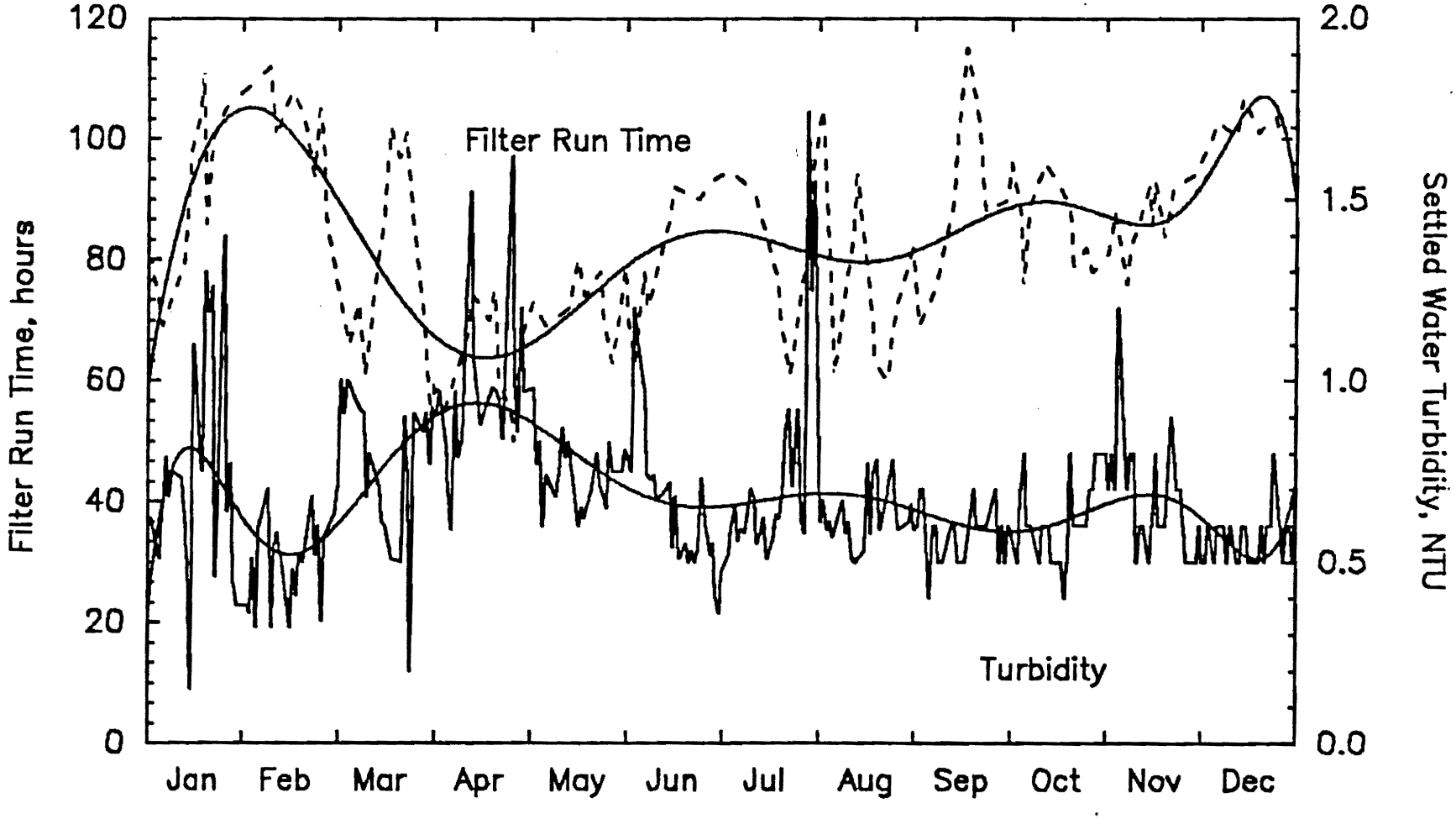

FIGURE 69. Length of Rapid Sand Filtration Runs Prior to Backwash and Settled Water Turbidity During 1988, Each with a Tenth-order Best-fit Regression Line to Indicate General Trends in the Data

Short periods of increased settled water turbidity also proved to have a negative impact on filter run time. The sharp increase in turbidity in the first two weeks of March resulted in a substantial decrease in filter run time from about 105 h to 65 h. When turbidity decreased to near its previous level, the length of a filter run increased to about 100 h. Near the end of March, turbidity increased to its highest value of the year and was sustained for about three months, resulting in a sustained period of decreased filter run times. As the turbidity decreased in mid-June, the length of a filter run subsequently increased.

From the above analysis of settled water turbidity and filtration operation, the general trends in filter run time can be related to solids loading on the filters, as indicated by settled water turbidity. Local decreases in filter run time can also be related to increased settled water turbidity. As discussed previously, some of these local areas of increased turbidity, such as occurred at the beginning of March, appeared to be the result of the use of recovered coagulant, although a definitive relationship could not be established.

A major impact of the use of recovered coagulant appears to be on the effectiveness of the sedimentation basin in the removal of particulate matter. A higher solids carryover to the filters was observed during some periods of recovered coagulant use, as indicated by settled water turbidity. These periods of increased settled water turbidity have a dramatic impact on the length of a filter run, causing a decrease in the length of a filter run and an increased backwash frequency. However, the filtration system maintained consistent finished water turbidity during use of commercial and recovered coagulants, as will be discussed later.

Finished Water Quality

Finished water quality during use of commercial and recovered coagulants can be compared based on pH, alkalinity, iron and manganese concentrations and turbidity, which were reported daily. Total trihalomethanes and other organic compounds were reported quarterly on grab samples taken from four random points in the distribution system.

Alkalinity and pH. As discussed previously, finished water pH is controlled by lime addition and should, therefore, not be impacted by the use of recovered coagulant. As indicated by data presented in Figure 70, finished water pH was maintained in the range of 7.6 to 8.2 through lime addition, indicating no impact from use of recovered coagulant. However, lime addition to maintain the target pH appears to be affected, as discussed previously.

As presented in Figure 70, finished water alkalinity ranged from about 140 to 160 mg/L as $CaCO_3$, with periods of dramatic fluctuations attributable to raw water quality. The perturbations in finished water alkalinity can be directly related to raw water alkalinity. Finished water alkalinity appears to follow the same trends as raw water alkalinity and shows no impact due to the use of recovered coagulant. For example, when water alkalinity decreased substantially in mid-January, finished water alkalinity subsequently decreased, although not as dramatically.

Manganese and Iron. As indicated earlier, raw water manganese concentrations are typically about 0.1 mg/L and as much as 0.75 mg/L can be expected to be added at the point of coagulant addition through the use of recovered coagulants. Finished water manganese, presented in Figure 71, indicates that the treatment system is adequate for the removal of a majority of this manganese. Finished water manganese concentrations ranged from zero to 0.03 mg/L, well below the previous potable-water manganese limit of 0.05 mg/L (USPHS 1965).

Finished water iron concentrations presented in Figure 72 were of the same magnitude as manganese, ranging from zero to 0.04 mg/L, significantly lower than the previous secondary potable water limit of 0.3 mg/L (USPHS 1965). As indicated previously, the high pH employed in the treatment process is amenable to oxidation of ferrous to ferric iron, thereby minimizing any impact of ferrous iron attributable to the recovered coagulant or in the raw water.

From the data presented above, it would appear that use of the recovered coagulant had no detrimental impact on finished water iron or manganese concentrations. The treatment scheme was adequate for oxidation and removal of any added manganese or ferrous iron attributable to the use of recovered

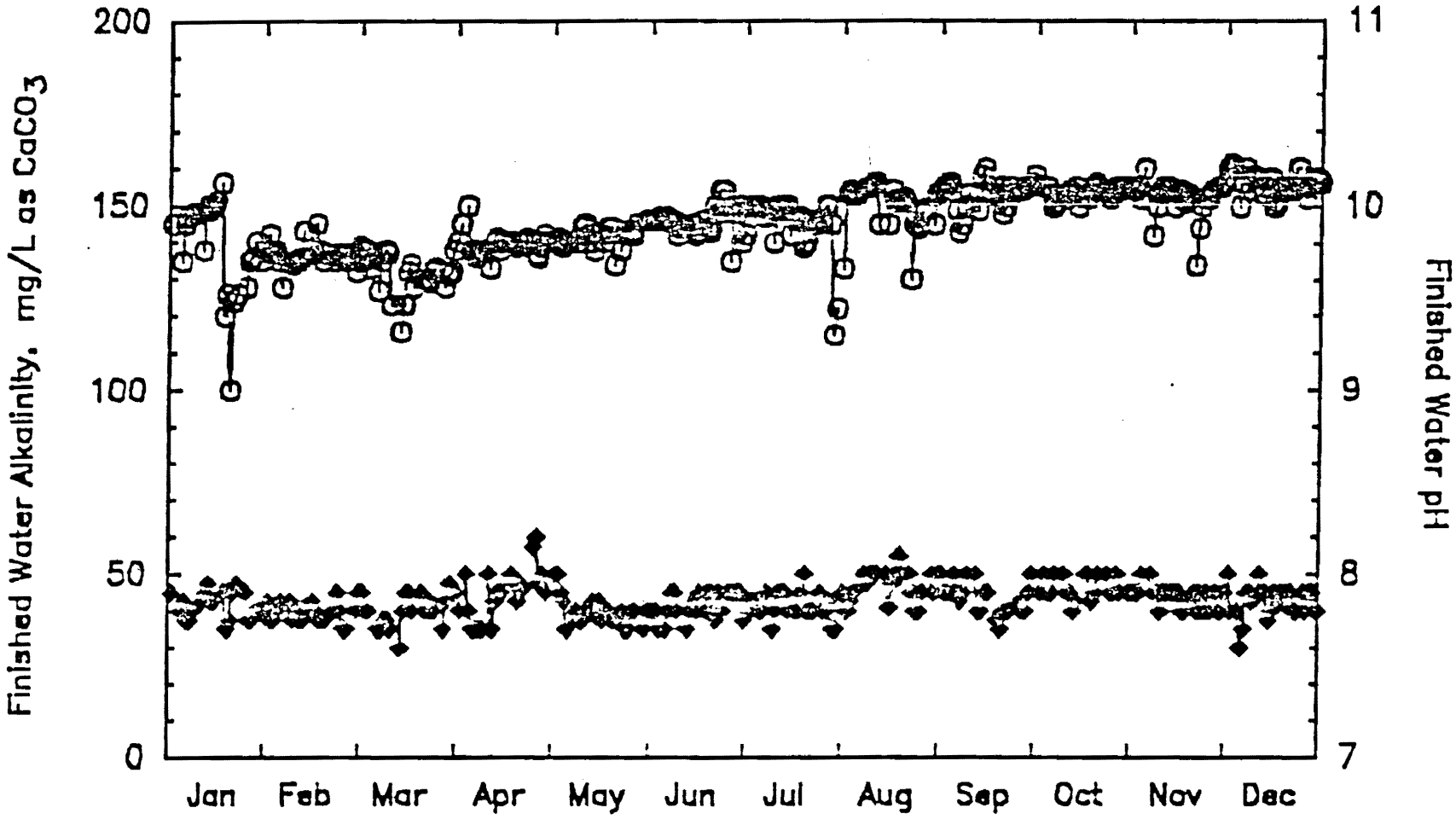

FIGURE 70. 1988 Finished Water Alkalinity and pH During Full-Scale Iron-Recovery Operations in Athens, TN

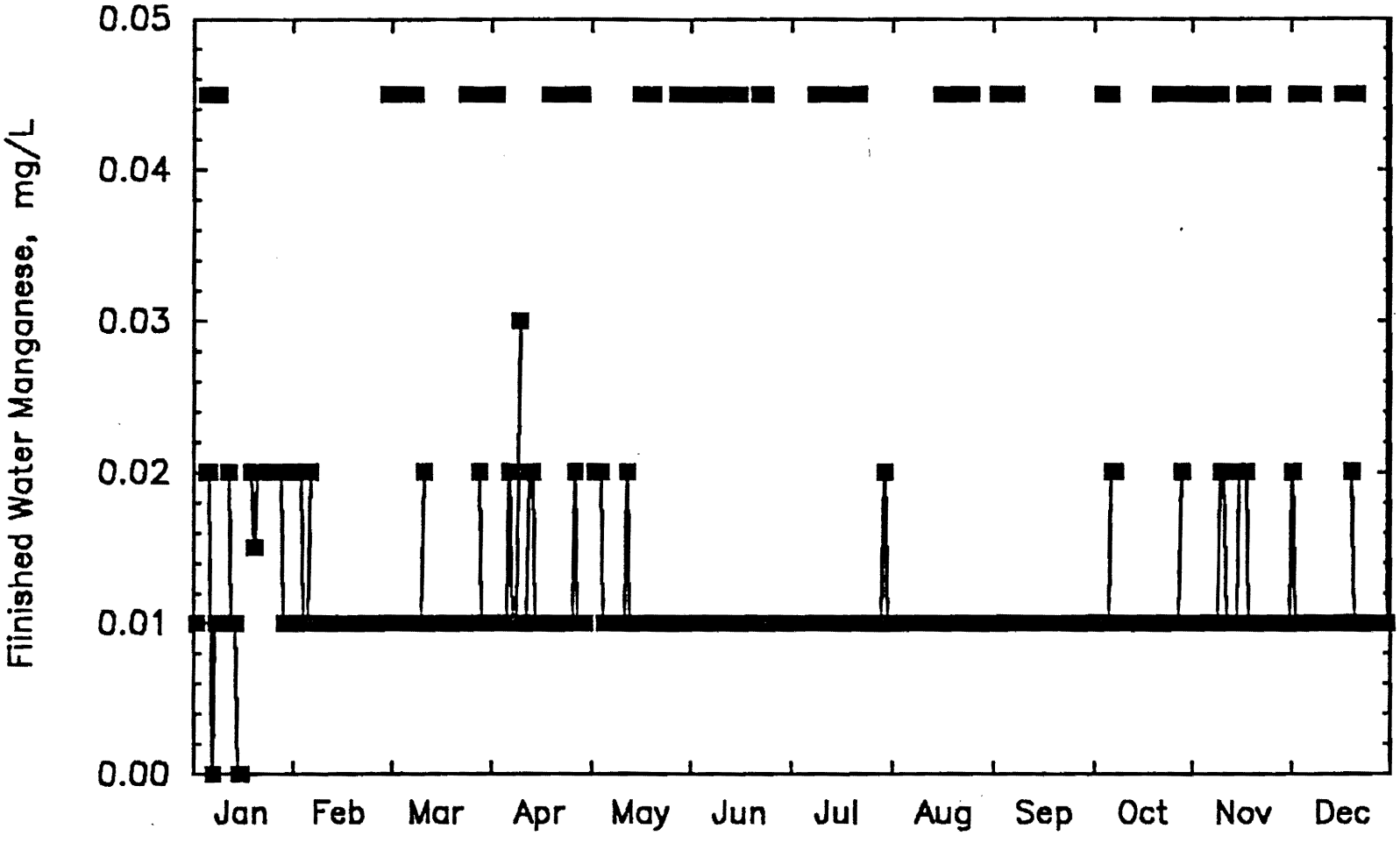

FIGURE 71. 1988 Finished Water Manganese Concentration and Indicated Periods of Recovered Coagulant Use

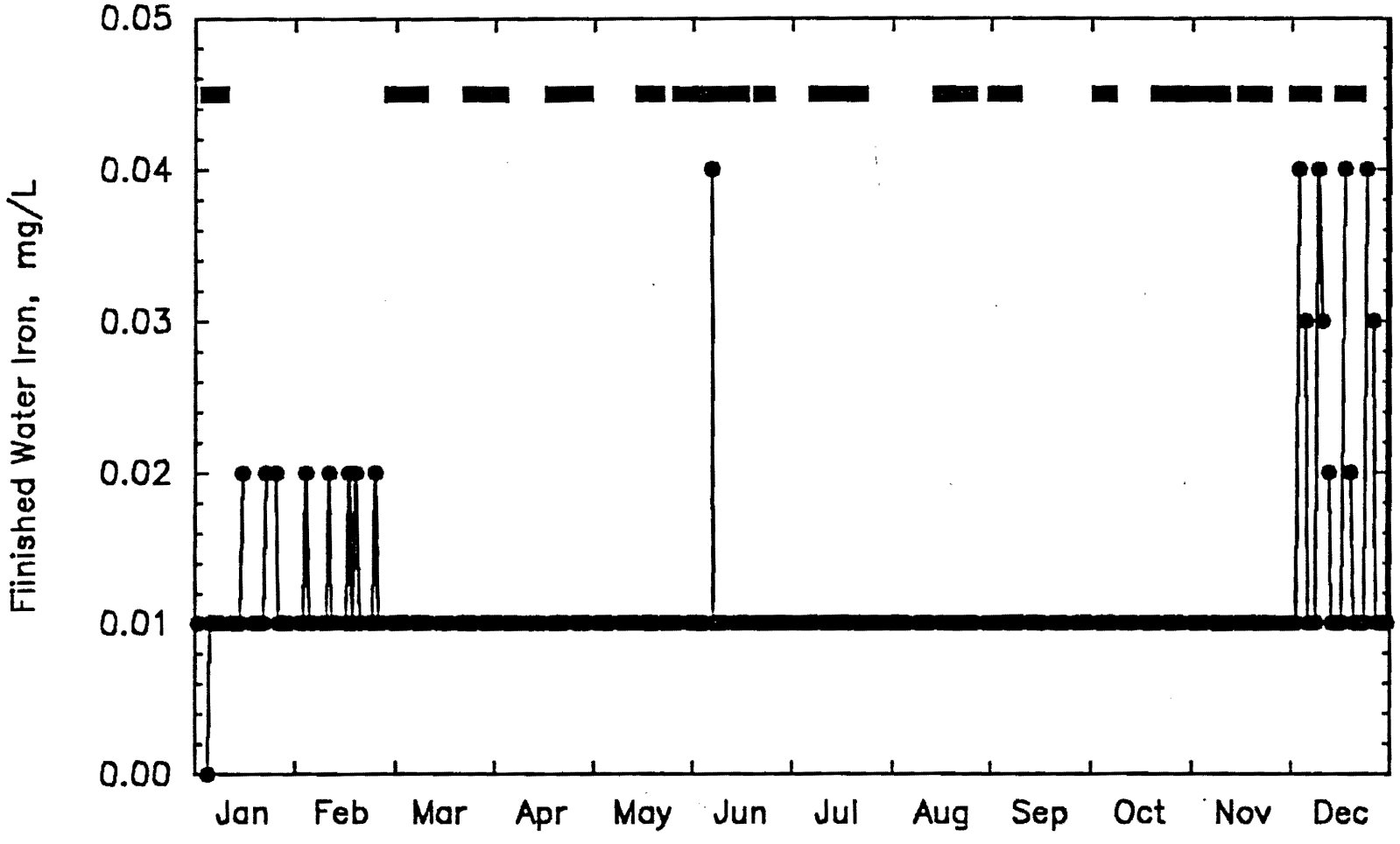

FIGURE 72. 1988 Finished Water Iron Concentrations and Indicated Periods of Recovered Coagulant Use

coagulants. Furthermore, finished water iron and manganese concentrations were always below acceptable limits of 0.3 mg/L and 0.05 mg/L, respectively.

Turbidity Removal. Turbidity is measured at three points in the treatment system; raw, settled and finished waters. Figure 73 presents these turbidity measurements on a log scale for 1988 to investigate turbidity removal through the plant. Turbidity is decreased from an average of about 7 NTU in the raw water to less than 1 NTU in the settled water. Following rapid sand filtration, finished water turbidity is typically less than 0.11 NTU.

Finished water turbidity is presented on an arithmetic scale in Figure 74 and ranged from 0.03 NTU to 0.11 NTU. These turbidity levels are significantly less than the current turbidity limit of 1.0 NTU (EPA 1972) and should easily meet the more stringent proposed standard of 0.5 NTU. These results indicate the efficiency of both the commercial and recovered coagulants in the agglomeration and destabilization of particles in the raw water. Although some degree of detrimental impact is observed in the efficiency of the sedimentation process during use of the recovered coagulants, results indicate that the rapid sand filtration system is adequate for the removal of particulate matter not removed in the sedimentation basin.

Organic Priority Pollutants and Regulated Inorganic Compounds. Organic priority pollutants and regulated metals are examined on a quarterly basis in accordance with established regulations. Although some sampling periods coincided with the use of recovered coagulants, these contaminant compounds are not specifically monitored during use of either coagulant to determine any impact from the use of recovered coagulants. However, data received from plant personnel indicate that all established MCLs are met in the finished water, as determined by quarterly grab samples taken in the distribution system.

In general, finished water quality was comparable during use of commercial and recovered coagulants. Use of the recovered coagulants does not appear to have a detrimental impact on finished water quality, as indicated by pH, alkalinity, turbidity and iron and manganese concentrations. Further investigation of other organic and inorganic contaminants is recommended to determine finished

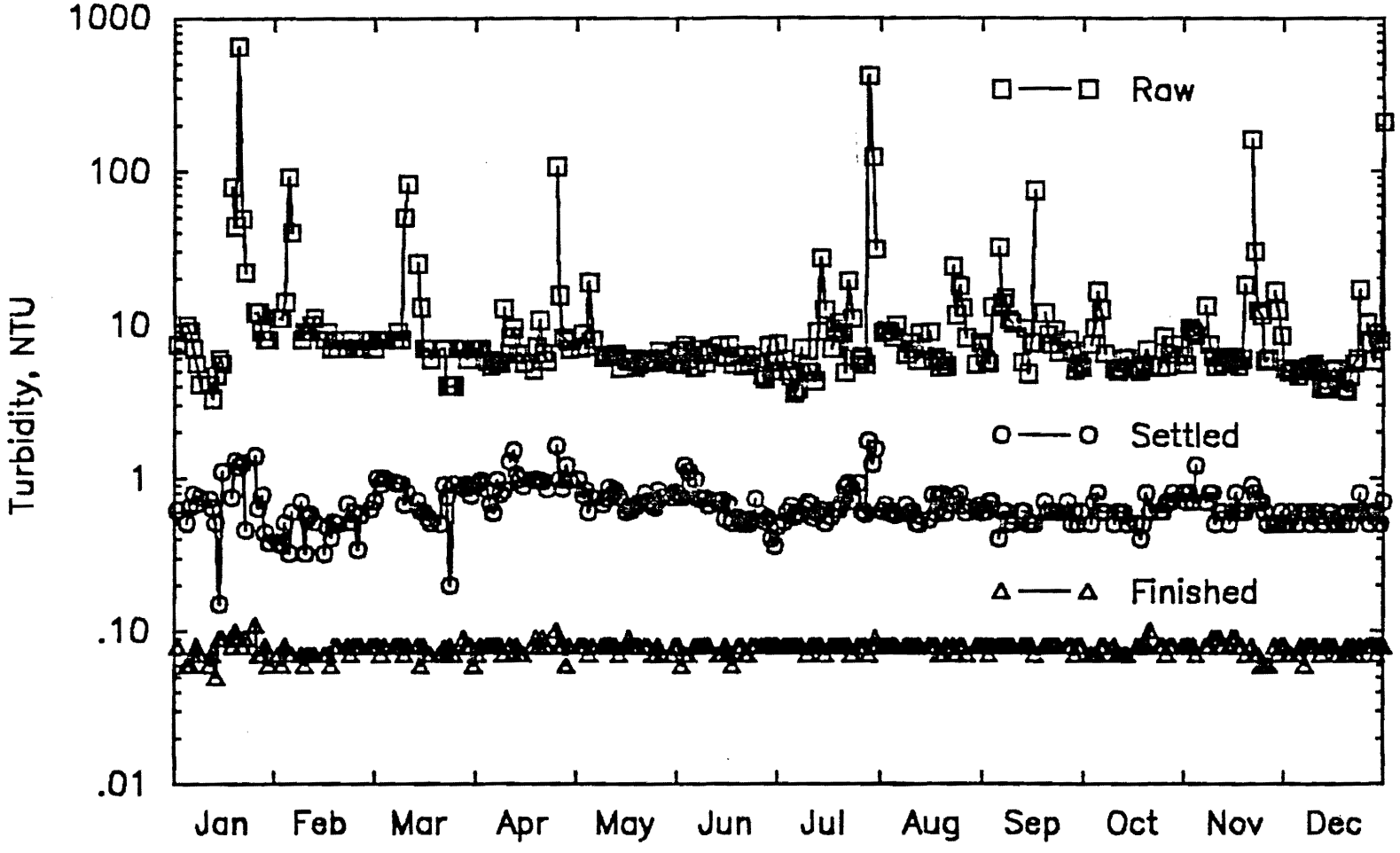

FIGURE 73. 1988 Raw, Settled and Finished Water Turbidity Profile on a Semi-log Scale

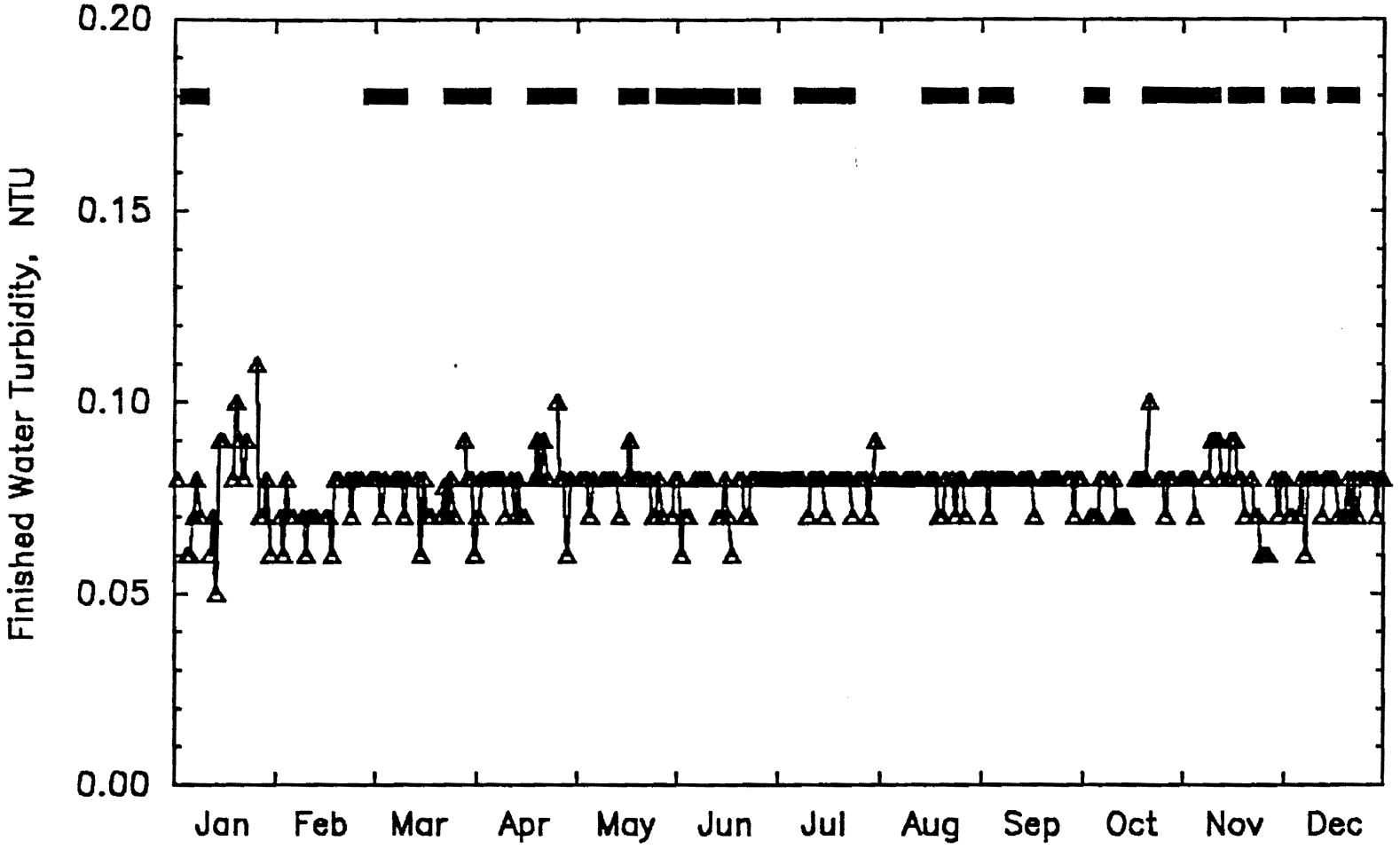

FIGURE 74. 1988 Finished Water Turbidity and Indicated Periods of Recovered Coagulant Use

water quality during use of the recovered iron coagulant product.

Economic Analysis of Coagulant Recovery at Athens, TN

An economic analysis of chemical operational costs with and without coagulant recovery was performed for 1988. As-purchased costs for commercial coagulant and actual costs of producing recovered coagulant using sulfuric acid and polymer were included in the analysis. Furthermore, average annual lime and chlorine addition data, presented earlier for periods of commercial and recovered coagulant use, were included in the analysis. Finally, total chemical costs for production of potable water during periods of commercial-coagulant and recovered-coagulant use were compared to determine the economic feasibility of coagulant recovery based on chemical costs alone.

A preliminary design analysis, conducted by an equipment manufacturer (White and Company 1983) was used to examine the predicted feasibility of coagulant recovery based on projected capital, operating and maintenance costs of coagulant recovery using vacuum-assisted dewatering compared to conventional sand drying beds and vacuum-assisted drying beds without coagulant recovery.

Economic Analysis of Chemical Operating Costs for 1988

The economical analysis for iron recovery at the Athens facility is based on chemical cost only for the year 1988. The analysis does not include capital costs since the capital expenses associated with implementation were not a factor in the annual costs of the plant. In addition, considerable effort was expended by plant personnel to refurbish a recovery system which had many design and built-in equipment limitations. As-purchased costs of chemicals used in the economical analysis were provided by the plant operator and are presented in Table 65. Chemical costs for production of recovered coagulant included those for sulfuric acid and polymer. In addition, incremental increases in lime and chlorine costs were included in the costs for production of potable water based on the observed increases in doses of these chemicals during use of recovered coagulant. All costs were normalized to flow treated during use of recovered coagulant and compared to costs of potable water production during use of commercial coagulant.

TABLE 65

Unit Costs of Chemicals Used in Potable-Water Treatment at the Athens, TN Plant (Camp 1989)

Chemical		Cost
Ferrifloc Coagulant	$Fe_2(SO_4)_3 \cdot 5H_2O$	\$0.079/lb $Fe_2(SO_4)_3 \cdot 5H_2O$
Lime	$Ca(OH)_2$	\$0.050/lb dry basis
Sulfuric Acid	Concentration 66° Be	\$0.048/lb H_2SO_4
Organic Polymer	Nonionic; Cyanamid 1986N	\$1.36/lb dry basis
Chlorine	Cl_2 gas	\$0.25/lb chlorine gas

During several brief periods in 1988, recovered coagulant was used for only a portion of a day or in conjunction with commercial coagulant. During these periods, the flow treated with recovered coagulant could not be accurately determined. Therefore, all chemical costs associated with the production of water for such periods were excluded from the economical analysis. Data for a total of 20 days in 1988 were involved, indicating the minimal impact of this exclusion.

The period of January through December 1988 was investigated in detail because recovered coagulant was used most intensively over this period. Approximately 35 percent of the total flow in 1988 was treated with recovered coagulant produced in 19 separate batches, including extractions made on a total of 39 days. In addition, only one volume of sludge produced on one day was dewatered without use of an acidic extraction. Furthermore, data collected over this period included iron concentrations for the recovered coagulants, allowing for investigation of coagulant mass application rates.

The dates of acidic extraction, chemical quantities used and costs and volume of recovered coagulant produced are presented in Table 66 for 1988. Acid extraction of sludge included addition of concentrated sulfuric acid to thickened sludges and polymer conditioning of the acidic suspensions prior to

TABLE 66

Economic Analysis of Chemical Costs for Production of Recovered Coagulants Used at the Athens, TN Plant in 1988

Date	Nonionic Polymer			Sulfuric Acid			Total Polymer and Acid Cost	Recovered Coagulant Volume	
	Volume		Cost	Mass		Cost			
	L	gal	$	kg	lb	$	$	m^3	gal
1987[1]									
12/23, 29	4.7	1.25	12.72	1,488	3,274	157.15	169.87	66.2	17,500
1988									
2/22, 25	5.7	1.50	15.26	2,400	5,279	253.39	268.65	83.3	22,000
3/21, 23	7.6	2.00	20.35	2,073	4,561	218.93	239.27	83.3	22,000
4/14, 15	5.7	1.50	15.26	2,531	5,569	267.31	282.57	88.9	23,500
5/10, 11	3.8	1.00	10.17	1,920	4,223	202.70	212.88	74.9	19,800
5/25, 26	5.7	1.50	15.26	2,837	6,242	299.62	314.88	79.5	21,000
6/2	3.8	1.00	10.17	2,170	4,774	229.15	239.32	55.6	14,700
6/6, 9	5.7	1.50	15.26	3,324	7,313	351.02	366.28	68.1	18,000
7/6, 12, 14	7.6	2.00	20.35	2,949	6,488	311.42	331.77	125.1	33,200
8/9, 10	5.7	1.50	15.26	2,545	5,600	268.80	284.06	88.6	23,400
8/30, 31	5.7	1.50	15.26	2,309	5,080	243.84	259.10	88.6	23,500
9/19, 20	5.7	1.50	15.26	3,262	7,176	344.45	359.71	61.7	16,300
10/3, 20	3.8	1.00	10.17	3,978	8,752	420.10	430.27	102.2	27,000
10/26, 27	5.7	1.50	15.26	2,594	5,707	273.94	289.20	74.9	19,800
11/1, 2, 3	5.7	1.50	15.26	2,997	6,594	316.51	331.77	115.4	30,500
11/14, 15	5.7	1.50	15.26	3,568	7,849	376.75	392.01	98.1	25,912
11/28, 12/1, 5	5.7	1.50	15.26	3,770	8,293	398.06	413.32	102.6	27,100
12/14, 15	5.7	1.50	15.26	3,651	8,033	385.58	400.84	73.4	19,400
Sum	103.1	27.25	277.21	44,511	97,924	4,700.35	5,761.21	1,584.4	418,612

[1]This batch of recovered coagulant was used in January 1988 and was therefore included in this analysis.

dewatering. Extractions were performed in batches which typically were conducted over 2-3 days.

As indicated previously, recovered coagulant was used in conjunction with commercial coagulant on 20 days in 1988. The volume of water treated with recovered coagulant on the given day could not be accurately determined during these periods. Therefore, the costs associated with commercial and recovered coagulant use on these occasions were excluded from the economic analysis presented. Using the volumetric application rate for recovered coagulants, the total volume of a given batch used during any period could be determined knowing the volume of water treated. When recovered coagulant was used for only a fraction of a day, the volume of recovered coagulant used was determined and its costs excluded from the economic analysis. The fraction of a given batch of recovered coagulant which was applied was determined by calculating the total volume of recovered coagulant used exclusively as a coagulant and dividing by the volume of recovered coagulant produced over that period. Commercial coagulant was treated in a similar manner, with the mass applied on that day being excluded from the analysis. Consequently, costs associated with production of potable water for 20 days during 1988 were excluded from the economic analysis presented. Additionally, the batch of recovered coagulant produced on September 19-20 was not used at the plant due to operational problems associated with its use, as discussed previously. Therefore costs associated with this batch were excluded from the economical analysis.

The cost of lime and chlorine addition was also included in the economic analysis since it was determined that application rates increased during use of recovered coagulant. Average lime and chlorine application rates increased 18.7 percent and 12.3 percent, respectively, during the use of recovered coagulant, as compared to that during use of commercial coagulant. Therefore, the annual average lime and chlorine application rates were employed in the economic analysis to accurately document all costs associated with recovered coagulant use.

The total flow treated with recovered coagulant during 1988 was 1.00×10^6 m^3 (265×10^6 gal), while the total flow treated with commercial coagulant was

1.86×10^6 m^3 (492×10^6 gal). That is, 35.0 percent of the total flow in 1988 was treated with recovered coagulant. All chemical costs were normalized to total flow treated as presented in Table 67. Production of recovered coagulant was less expensive by $\$0.824/10^3$ m^3 ($3.12/10^6$ gal) of water treated, than the use of commercial coagulant. Lime and chlorine costs were higher during use of recovered coagulant, however, by $\$0.48/10^3$ m^3 ($\$1.81/10^6$ gal) and $\$0.27/10^3$ m^3 ($\$1.04/10^6$ gal), respectively, for a total of $\$0.75/10^3$ m^3 ($\$2.85/10^6$ gal). Although costs associated with lime and chlorine application were higher during use of recovered coagulant, the total chemical costs during use of commercial and recovered coagulants were essentially equal at $\$10.65/10^3$ m^3 (40.30 $\$/10^6$ gal) and $\$10.58/10^3$ m^3 (40.03 $\$/10^6$ gal), respectively.

TABLE 67

Chemical Costs for Commercial and Recovered Coagulants, Lime and Chlorine During Full-Scale Coagulant Recovery Operations at Athens, TN

Chemical	Commercial Coagulant Use			Recovered Coagulant Use		
	Cost	Unit Cost		Cost	Unit Cost	
	$	$/10^6 gal	$/10^3-m^3	$	$/10^6 gal	$/10^3-m^3
Coagulant	10,907.77	22.15	5.85	5,041.22	19.03	5.03
Lime	4,768.09	9.68	2.56	3,045.32	11.49	3.04
Chlorine						
Pre-	2,043.17	4.15	1.10	1,276.25	4.82	1.27
Post-	2,125.31	4.32	1.14	1,243.10	4.69	1.24
Total Cost	19,844.34	40.30	10.65	10,605.89	40.03	10.58

The data presented above indicate that coagulant recovery is economically feasible at this site based on total chemical costs for drinking water production. However, capital costs associated with the acid reactor and acid

resistant equipment and materials have not been considered in the above analysis. Furthermore, economic savings associated with the reduction in sludge volume and mass for disposal have not been considered. Inadequate information is available on the above issues to enable an in-depth study, however, preliminary design information and cost analysis for the coagulant recovery system is presented in the following section.

Preliminary Analysis of Capital, Operating and Maintenance Costs

A preliminary design analysis was conducted in 1983 to investigate the economic feasibility of construction and operation of a coagulant recovery system at the Athens site (White and Company 1983). The preliminary design evaluated conventional sand drying beds, vacuum-assisted drying beds with polymer addition and vacuum assisted beds employing coagulant recovery.

A maximum design flow of 4.0 mgd was utilized assuming 24 h/d, 7 d/wk operation, with average chemical applications of 31.8 mg/L of ferric sulfate and 26.8 mg/L of lime. The total solids production used for design of the sludge handling system was 87.7 kg/m^3 (730 lb/10^6 gal), or 484,545 kg/y (1,066,000 lb/y). As presented in Table 68, the conventional sand drying bed system (SDB) was designed for 3 cycles/y, requiring a total area of 11705 m^2 (126,000 ft^2). The vacuum bed with polymer addition (VB) and the vacuum bed utilizing coagulant recovery (VB-CR) were designed for 21-h cycles, with required bed areas of 371.6 m^2 (4000 ft^2) and 139.3 m^2 (1500 ft^2), respectively.

Capital costs for the SDB, VB and VB-CR systems were $1,078,000, $350,000 and $360,000, respectively. Although the VB-CR system required 60 percent less bed area than the VB system, other capital costs were involved with the VB-CR system, such as an acid-contact basin, an acid storage tank, a recovered coagulant storage tank and acid resistant piping and equipment.

Operation and maintenance costs are presented in Table 69 for each of the systems investigated. Based on a commercial coagulant dose of 31.8 mg/L as $Fe_2(SO_4)_3 \cdot 5H_2O$ the annual commercial coagulant cost would be $81,000 for the SDB and VB system, while the VB-CR system would require only $20,000 annually

TABLE 68

Preliminary Design Criteria and Capital Costs Associated with Conventional Beds (SDB), Vacuum Assisted Beds with Polymer Conditioning (VB) and Vacuum Assisted Beds with Coagulant Recovery (VB-CR) (White and Company 1983)

	Conventional Sand Beds (SDB)	Vacuum Beds with Polymer (VB)	Vacuum Beds with Coagulant Recovery (VB-CR)
Raw Sludge Solids, kg/y	2.345×10^6	2.345×10^6	2.345×10^6
Operational Cycles	3 cycles/y	21-h/cycle	21-h/cycle
Drying Bed Area, m^2 (ft^2)	11,706 (126,000)	372 (4,000)	139 (1,500)
Capital Costs, $	1,078,000	350,000	360,000

assuming 75 percent recycle of coagulant. However, an additional cost of $11,000 annually would be incurred for the VB-CR system from use of sulfuric acid in the coagulant recovery process. Polymer costs totalled $8000/y and $4000/y for the VB and VB-CR systems, respectively, while both would incur a power cost of $2000/y. Cake hauling and disposal costs for the SDB system were the lowest at $4300/y, followed closely by VB-CR at $4500/y. Total annual operating and maintenance costs of $134,000, $137,100 and $79,500, respectively for the SDB, VB and VB-CR systems, resulted from the above analysis.

Total annual costs were developed based on the previous capital operating and maintenance costs, assuming financing over a 30-year period. As presented in Table 70, the vacuum assisted bed utilizing coagulant recovery proved to be the least expensive at an annual cost of $126,500, followed by VB at $183,100 and SDB at $274,000, 45 percent and 120 percent higher than VB-CR, respectively.

Based on the above analysis of capital, operating and maintenance costs, the coagulant recovery system for solids conditioning and vacuum assisted drying

TABLE 69

Preliminary Operation and Maintenance Costs for Conventional Sand Drying Beds, Vacuum Assisted Drying Beds with Polymer and Vacuum Assisted Drying Beds with Coagulant Recovery (White and Company 1983)

	Annual Costs, $/y		
	Conventional Sand Beds	Vacuum Beds with Polymer	Vacuum Beds with Coagulant Recovery
Commercial Coagulant	81,000	81,000	20,000 (75% recycle)
Sulfuric Acid	0	0	11,000
Polymer	0	8,000	4,000
Other Chemicals	11,500	12,500	9,000
Power	0	2,000	2,000
Personnel	20,000	15,000	20,000
Bed Maintenance	18,000	10,000	9,000
Cake Hauling and Disposal	4,300	8,600	4,500
Total Annual Operation and Maintenance	134,800	137,000	79,500

TABLE 70

Total Annual Cost Estimate for Construction, Financing and Operating and Maintenance Costs Based on 30-Year Financing of Conventional Sand Drying Beds, Vacuum Drying Beds with Polymer and Vacuum Drying Beds with Coagulant Recovery (White and Company 1983)

	Annual Costs, $/yr		
	Conventional Sand Beds	Vacuum Beds with Polymer	Vacuum Beds with Coagulant Recovery
Principal and Interest[1]	140,000	46,000	47,000
Operation and Maintenance	134,800	137,100	79,500
Total Annual Operation and Maintenance	274,800	183,100	126,500

[1]Based on city financing over a 30-year period

beds proved to be considerably less expensive than the other systems investigated. Furthermore, the required bed area was significantly lower for the VB-CR system and could be attractive at a site with limited area.

Discussion of Results

Coagulant recovery has been successfully practiced at the Athens, TN facility since October 1985. A total of 28 batches of recovered coagulant were produced and re-used as primary coagulant at the plant, with about 35 percent of the water is being treated with recovered coagulant in 1988. The recovered product was capable of producing a finished water equivalent to that using commercial coagulant, as indicated by pH, alkalinity, turbidity, and iron and manganese concentrations in the finished water. However, increased lime and chlorine application rates were required during use of recovered coagulant to maintain the pH and provide for increased oxidant demand.

Use of the recovered product decreased the efficiency of the sedimentation system, as indicated by increased solids loading on the filters. This resulted in a decrease in the length of a filter run prior to backwash, but had no apparent impact on finished water quality.

Based on chemical costs alone in the period of January through December 1988, the cost of drinking water production was equivalent during use of commercial and recovered coagulants at about $\$10.57/10^3$ m^3 ($\$40/10^6$ gal). The costs of the increased chemical application was offset by the lower cost of recovered coagulant production as compared to commercial coagulant.

A preliminary design analysis of capital, operating and maintenance costs indicated that coagulant recovery for solids conditioning and vacuum assisted drying bed dewatering was considerably less expensive than conventional sand drying beds or vacuum-assisted beds without coagulant recovery. The required area for the vacuum beds utilizing coagulant recovery was substantially less than for the conventional system, making coagulant recovery and vacuum assisted dewatering an attractive option for facilities with limited land area.

AMERICAN WATER WORKS SERVICE COMPANY

The American Water Works Service Company, Inc. (1987) investigated numerous options for conditioning and dewatering of water treatment plant sludges for a comparative analysis of capital costs, operating and maintenance costs and 20-year present worth of each system at various solids loadings. Sand drying beds, Wedgewater filter beds, Dehydro dewatering systems, coagulant recovery and belt filter press dewatering were compared based on economic considerations and physical parameters, such as land area requirements and total volume of sludge produced. Pilot- and full-scale evaluations of each system provided the design criteria utilized in calculating the physical and economic parameters presented.

The coagulant recovery system involved six major components including a sulfuric-acid storage and pumping system, a reaction tank for acid-sludge contact, a polymer preparation and metering system, a vacuum-assisted dewatering bed, vacuum generation equipment and controls and a dewatering bed filtrate collection and pumping system. The economic analysis presented was based on capital, operating and maintenance costs of the conditioning and dewatering system alone, and did not take into consideration economic implications for water treatment. That is, no economic savings associated with the decreased use of commercial coagulant were incorporated into the economic analysis for coagulant recovery.

Land area requirements, total cake production, capital costs, operating and maintenance costs and 20-year present worth of each system investigated are presented in Table 71 as a function of water treatment plant solids production. Land area requirements were considerably lower for the coagulant recovery system at lower solids loadings, e.g., up to 955 kg/d (2100 lb/d), while at a solids loading of 3409 kg/d (7500 lb/d) of dry solids belt filter press dewatering had the lowest required land area. The total volume of wet sludge cake produced was always lowest for the coagulant recovery system. A total of 83 m^3/y (2993 ft^3/y) of wet sludge solids would require disposal from a water treatment plant producing 136 kg/d (300 lb/d) of dry solids, while 1972 m^3/y (69,493 ft^3/y) of sludge would require disposal at a plant producing 3409 kg/d (7500 lb/d).

TABLE 71

Economic Analysis of Sand Drying Beds, Wedgewater Filter Beds, Dehydro Sludge Dewatering, Coagulant Recovery and Belt Filter Press Systems for Water Treatment Plant Sludge Conditioning and Dewatering (American Water Works Service Company 1987)

	Sand Drying Beds	Wedgewater Filter Beds	Dehydro System	Coagulant Recovery System	Belt Filter Press
136 kg/d DS* (300 lb/d)					
Land Area, m^2 (ft^2)	844 (9,070)	163 (1,750)	223 (2,400)	13 (135)	140 (1,500)
Cake Production, m^3/y (ft^3/y)	233 (8,228)	173 (6,107)	247 (8,703)	85 (2,993)	201 (7,093)
Capital Costs, $1000	212	127	298	154	412
O&M Costs, $1000/y	11.7	9.5	17.2	22.6	22.6
20-y Present Worth, $1000	584	400	831	688	1,117
455 kg/d DS (1,000 lb/d)					
Land Area, m^2 (ft^2)	2,698 (29,000)	326 (3,500)	651 (7,000)	40 (425)	140 (1,500)
Cake Production, m^3 (ft^3/y)	761 (26,820)	607 (21,378)	786 (27,697)	263 (9,235)	649 (22,860)
Capital Costs, $1000	649	198	589	201	532
O&M Costs, $1000/y	35	35	52	44	35
20-y Present Worth, $1000	1,777	1,023	2,007	1,174	1,568
955 kg/d DS (2,100 lb/d)					
Land Area, m^2 (ft^2)	5,713 (61,415)	707 (7,600)	1,414 (15,200)	85 (909)	140 (1,500)
Cake Production, m^3 (ft^3/y)	1,581 (55,715)	1,292 (45,535)	1,674 (58,993)	562 (19,802)	1,375 (48,464)
Capital Costs, $1000	1,366	403	2,370	397	532
O&M Costs, $1000/y	73	76	113	79	72
20-y Present Worth, $1000	3,717	2,190	6,128	2,182	2,327
3,409 kg/DS (7,500 lb/d)					
Land Area, m^2 (ft^2)	20,235 (217,525)	2,456 (26,400)	4,912 (52,800)	525 (5,640)	140 (1,500)
Cake Production, m^3 (ft^3/y)	5,599 (197,341)	4,571 (161,104)	5,922 (208,707)	1,972 (69,493)	4,862 (171,371)
Capital Costs, 1000$	4,800	1,174	8,019	950	856
O&M Costs, 1000$/y	249	267	396	193	216
20-y Present Worth, $1000	12,947	7,283	21,028	5,348	5,813

*DS = Dry Sludge Solids

Capital costs for the coagulant recovery system were increasingly favorable as compared to the other systems investigated, ranging from $154,000 at 136 kg/d (300 lb/d) of dry solids to $950,000 at 3409 kg/d (7500 lb/d) of dry solids. The belt filter press dewatering system required the lowest capital investment for the 3409 kg/d (7500 lb/d) mass flow at a cost of $856,000. Operation and maintenance costs for the coagulant recovery system were the highest of the systems compared at 136 kg/d (300 lb/d) of dry solids, but became increasingly favorable as the solids loading increased to 3409 kg/d (7500 lb/d) where coagulant recovery exhibited the lowest operation and maintenance costs of the systems investigated. As indicated by the trends noted above, the coagulant recovery system becomes more economically feasible as the plant solids production increased.

The present worth of the coagulant recovery system indicated that as the solids loading to the dewatering system increased, coagulant recovery becomes more cost effective. That is, at 136 kg/d (300 lb/d) loading of dry solids the coagulant recovery system has a relatively high present worth of $688,000, compared to the lowest present worth of $400,000 for the Wedgewater filter bed system. As the solids loading increases to 3409 kg/d (7500 lb/d) of dry solids, the coagulant recovery system has the lowest present worth value at $5,348,000, compared to the next highest present worth of $5,813,000 for the belt filter press dewatering system.

In general, the economic analysis indicated the relative economic efficiency of the coagulant recovery system in comparison to four other systems investigated. As the solids loading to the dewatering systems increased, the relative costs of the coagulant recovery system decreased. From the above data base, it appeared that coagulant recovery is more economically feasible at sites with higher solids loadings. Furthermore, when land area is limited, coagulant recovery provides an attractive alternative, requiring considerably less land area than the other systems evaluated. The final volume of sludge for disposal was always dramatically lower for the coagulant recovery system due to the dissolution of the voluminous aluminum hydroxide solids in the acid extraction process.

SPARTANBURG, SC: PRELIMINARY INVESTIGATIONS ON COAGULANT RECOVERY

Jordan, Jones and Goulding, Inc. (1988) conducted a pilot-scale study of solids handling at the R. B. Simms Filtration Plant in Spartanburg, SC for determination of design parameters to conduct a preliminary cost analysis. The proposed system evaluated consisted of a washwater handling system, sedimentation basin solids collection system, sludge thickeners and sand drying beds. A comparative cost analysis was conducted for the proposed system employing coagulant recovery prior to sludge application to drying beds.

Historical plant records were utilized for estimating solids loadings to the solids handling facility. Using the maximum monthly solids loading, the design calculations were based on a solids loading of 14.1 g/m^3 (117.299 $lbs/10^6$ gal) of water treated. Analysis of sludge aluminum content revealed that 12.2 percent of the sludge solids were composed of aluminum, which is equivalent to 59.6 percent of the solids if the aluminum is in the form of $Al(OH)_3 \cdot 3H_2O$. Based on studies of sulfuric acid extraction of aluminum and residual solids separation, it was determined that 36 percent of the total aluminum present in the sludge could be recovered for reuse as a coagulant. Polymer application was required for adequate dewatering performance in pilot-scale drying bed tests. A 40 percent reduction in the mass of sludge solids applied to the drying beds would be experienced due to dissolution of aluminum sludge solids and a corresponding 40 percent reduction in required drying bed area would be realized.

Based on the above information and a more detailed design of the solids dewatering system with and without coagulant recovery, an economic analysis was performed, the results of which are summarized in Table 72. Capital costs were $3,204,166 and $3,412,384 without and with coagulant recovery, or an increased cost of $208,218 (6.5 percent) for the coagulant recovery system. Increased chemical costs for polymer, acid and caustic would be required for coagulant recovery operation. However, a savings of $29,923/y would be associated with recovery of 36 percent of the commercial coagulant for reuse as recovered coagulant. The coagulant recovery system had higher labor costs for operation and maintenance of the system, but had considerably lower labor

TABLE 72

Solids Handling and Alum Recovery Economic Summary Based on Pilot-Scale Evaluation of Coagulant Recovery at Spartanburg, SC
(Jordan, Jones and Goulding, Inc. 1988)

Parameter	Solids Handling	Solids Handling with Alum Recovery	Additional Cost
Capital Cost	$3,204,166	$3,412,384	$208,218
Debt Service	308,339	328,376	20,037
Chemicals			
Coagulant	83,119	53,196	(29,923)
Polymer (Thickening)	7,079	7,079	0
Polymer (Decant)	0	4,831	4,831
Acid	0	15,090	15,090
Caustic	0	11,983	11,983
Supplies/Parts	1,500	7,500	6,000
Labor			
Alum Rec. Operation	0	2,520	2,520
Alum Rec. Maint.	0	3,157	3,157
Thick./Recyc. Oper.	2,512	2,512	0
Thick./Recyc. Maint.	789	789	0
Draw Beds	6,138	4,582	(1,556)
Clean Beds	58,797	35,278	(23,519)
Landfill			
Excavation/Backfill	6,282	3,769	(2,513)
Haul (Oper. Cost)	485	291	(194)
Electricity	8,231	10,157	1,926
Total Annual Cost	$ 483,271	$ 491,110	$ 7,839

costs for drying bed maintenance due to a 40 percent decrease in the area requiring service.

Costs associated with landfilling the residual sludge solids were 40 percent lower using coagulant recovery, resulting in an annual sludge disposal savings of $2707/y. Compared to typical sludge disposal costs associated with landfilling water treatment sludges, these costs are minimal due to the limited hauling distance and landfill ownership. The sludge is disposed of in a landfill owned and operated by the water authority near the site of the water treatment plant. The hauling distance is only 0.37 km (1.5 miles) and no tipping fee is required for disposal of sludge solids. The landfill site is pre-existing, requiring no land purchase or developmental costs. Therefore, costs associated with landfilling the residual sludge solids at this facility are low, as compared to those at a facility that is required to, for example, haul and dispose of sludge solids at a municipal landfill.

The total annual costs associated with the solids handling facility was $483,271/y without coagulant recovery and $491,110/y with coagulant recovery, an increased cost of $7839/y (1.6 percent) associated with use of the coagulant recovery system. Based on the above results, the coagulant recovery system was not recommended for full-scale installation at the Spartanburg facility, but provisions will be made for potential installation in the future. Installation of a coagulant recovery pilot-plant was recommended to further pursue the investigation into coagulant recovery based on the actual characteristics of the sludge solids produced from the new solids handling facility.

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